

likely the result of DPE remediation (Dwells et al., 2007).

### 5.5.2 Isotopic fractionation as a proxy indicator of biodegradation

The last few years have seen considerable interest in the use of compound specific isotope analysis (CSIA) as a tool to document biodegradation of MTBE in the subsurface (*Kolhatkar et al., 2002; Kuder et al., 2005; Wilson et al., 2005a; Zwank et al., 2005; USEPA 2005*). Simply put, the methodology stems from the principle that microorganisms preferentially biodegrade the chemical with lighter isotope (for example  $^{12}\text{C}$ , which is more common), resulting in enrichment in the heavier isotopes (e.g.,  $^{13}\text{C}$ , which is less common), a process referred to as isotopic fractionation. For MTBE, CSIA was seen as a powerful tool, making it possible to “unequivocally identify and measure anaerobic biodegradation” (*USEPA 2005*). In applying this methodology it was assumed that other processes that can fractionate compounds in the subsurface, such as volatilization, adsorption, diffusion, etc...have a negligible isotopic effect compared to biodegradation. Recent research however, indicates otherwise: “Use of CSIA is no silver bullet and will be most useful and cost effective when applied within the context of the hydrological, geological, geochemical and microbiological parameters at the site. The factors that affect contaminant transport and degradation over time as well as space must be identified and evaluated.” (*USEPA, 2008*).

Here, I discuss:

- The limitations of CSIA application to field sites as an indicator of MTBE biodegradation, given recent studies that show that other physical and chemical processes can in fact contribute to isotopic fractionation.
- The published reports on the use of CSIA to document anaerobic MTBE biodegradation, especially with regards to Orange County, California, and the related MTBE “natural” attenuation research performed by the USEPA (2005).

### 5.5.2.1 Limitations in CSIA application at field sites

A fundamental limitation in the application of CSIA to groundwater systems stems from both the known and the unknown potential for other physical and chemical processes to produce similar isotopic fractionation signals to those produced by biodegradation. The study of the effects of physical and chemical processes on isotopic fractionation is an evolving and active field of research (*Pernaton et al., 1996; Prinzhofner and Pernaton, 1997; Poulsen et al., 1997; Harrington et al., 1999; Zhang and Kroos, 2001; Ellis et al., 2002; Schloemer and Kroos, 2004; Richter et al., 2006; LaBolle et al., 2006; Kuder et al., 2007; Van Breukelen 2007; Bouchard et al., 2008; Kuder et al., 2009*). In a recent publication, USEPA (2008) noted: “Because CSIA is a new approach, there are no widely accepted standards for accuracy, precision and sensitivity, and no established approaches to document accuracy, precision, sensitivity and representativeness” (USEPA 2008).

For example, recently published results (*LaBolle et al., 2006*) indicate that diffusion in water can lead to isotopic enrichment effects similar to those attributed to aerobic biodegradation. Figure 5.5 compares the computed isotopic enrichment of  $^{13}\text{C}$  MTBE by diffusion in an alluvial aquifer with that measured during MTBE biodegradation in laboratory microcosms showing how they may produce similar effects (*LaBolle et al., 2006*).

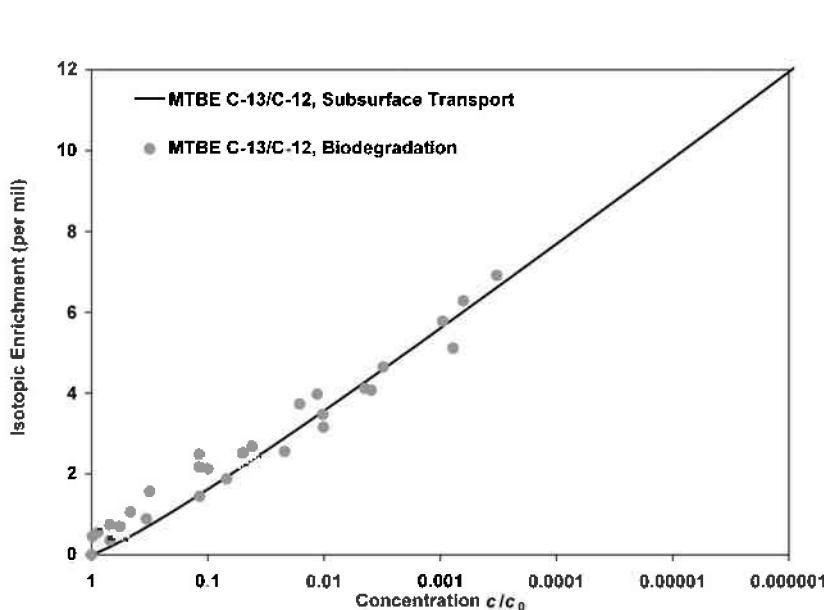


Figure 5.5. Computed  $^{13}\text{C}$  MTBE isotopic enrichment resulting from transport in a thin alluvial aquifer bounded by aquitards is compared with measured isotopic fractionation resulting from aerobic biodegradation of MTBE (*Hunkeler, 2001*). Results show the potential for diffusion to confound efforts to assess biodegradation using CSIA in the field.

Research in the late 1990's demonstrated that volatilization has a significant effect on the isotopic fractionation of VOCs. A seminal compendium of papers on isotopic fraction appeared in the journal Organic Chemistry in 1999 (Vol. 30). The introductory note to those publications called attention to the role of volatilization:

“A reasonable conclusion to draw from this set of papers is that while physical fractionation of carbon and chlorine isotopes may be small under vapor/liquid equilibrium conditions, such isotopic fractionation may be magnified at large degrees of evaporation. Such potential Rayleigh reservoir effects must therefore be taken into account in natural or engineered systems.” (*Abrajano and Lollar, 1999*).

However, that advice and its importance, especially with regards to remediation, were largely ignored - volatilization was generally assumed to have negligible effects on isotopic fractionation, until recently. Current research on the volatilization of MTBE indicates significant isotopic effects (*Philp and Kuder, 2006; Kuder et al., 2007*). Based on laboratory studies, Kuder et al., (2009) concluded: “Experimental results on MTBE behavior in diffusive volatilization and dynamic vapor extraction show measurable changes in the isotope ratios of the MTBE remaining in the aqueous or nonaqueous phase liquid (NAPL) matrix”.

Even so, the simplified bench-top laboratory models used by Kuder et al., (2009) only touched on the possible extent of this phenomenon at field sites, which span tens to hundreds of feet. Remediation processes at field sites, such as dual phase extraction (DPE), air sparging (AS) and soil vapor extraction (SVE) involve the four-phase transfer of MTBE between air, water, soil and free-phase gasoline, as well as the transport of groundwater and gasoline vapors over tens to hundreds of feet, and the redistribution of gasoline from a vapor to a free-phase in the vicinity of the extraction well. Isotopic fractionation will likely occur in all of these mass transfer processes (*Eweis et al., 2007*).

Abiotic transformation of MTBE to TBA can also contribute to isotopic fractionation. For example, a recent study (*Elsner et al., 2007*) reported significant isotopic fractionation of MTBE resulting from acid hydrolysis (abiotic degradation). As discussed earlier, MTBE may also be reduced abiotically to TBA under anaerobic (methanogenic) conditions (*Bradley et al., 2006*). Given that this is relatively new research, the effect of abiotic MTBE reduction on isotopic fractionation, to my knowledge, is unknown at this point in time. Nevertheless, studies on other contaminants indicate that isotopic fractionation due to reductive processes can be significant. VanStone et al. (2004) for example, reported significant enrichment in  $^{13}\text{C}$  in chlorinated ethylenes as a result of abiotic reductive dechlorination. Similarly, Bill et al. (2001) reported that lighter isotopes reacted preferentially in abiotic reductive dechlorination of TCE resulting in  $^{13}\text{C}$  enrichment. As a result, USEPA (2008) notes: “The relative importance of biodegradation versus processes of abiotic degradation at the site must be considered. In the past few years, the principles of Rayleigh controlled isotope fractionation of organic contaminants in ground water have been shown to apply to abiotic degradation as well as biodegradation”

Further work is needed therefore, both in the laboratory and in the field, to evaluate the effects of remediation and abiotic transformation processes on isotopic fractionation. To my knowledge, the effects of SVE and DPE, as well as other remediation processes and hydrogeologic factors, on the isotopic fractionation of MTBE have not been studied under field conditions.

### ***5.5.2.2 The USEPA (2005) Report on “Monitored Natural Attenuation of MTBE”***

The USEPA document “Monitored Natural Attenuation of MTBE as a Risk Management Option at Leaking Underground Storage Tank Sites” (*USEPA 2005*) relied heavily on the use of isotopic fractionation as an indicator of anaerobic MTBE biodegradation in the subsurface asserting that: “This makes it possible for the first time to unequivocally identify and measure anaerobic biodegradation of MTBE at field Scale” (*USEPA 2005*). The EPA document (*USEPA 2005*) relied on the work of J.T. Wilson (*USEPA*) and others and evolved out of a need to interpret data from MTBE-contaminated sites where relatively high concentration ratios of TBA to MTBE were observed in monitoring wells. Theoretically, high TBA to MTBE could result from MTBE biodegradation to TBA.

In this section, I will discuss the research findings of Wilson et al. (2005a) and Kuder et al. (2005) and the data that they collected from thirteen Orange County, California MTBE-contaminated sites, and which formed the basis for *USEPA (2005)*<sup>22</sup>. I will also provide details from the recent analysis and review of this research by Eweis et al. (2007). The research that led to the publication of Eweis et al. (2007) was conducted with the cooperation of Dr. J.T. Wilson (*USEPA*) who provided my colleagues and I with details regarding the analytical results from the 13 Orange County sites that he studied. He did not participate in the actual analysis. We also gathered additional publicly available data from the California State Water Resources Control Board Geotracker database, including quarterly reports that describe the investigation and remediation activities performed at the 13 Orange county MTBE-contaminated sites discussed in *USEPA (2005)* (see Table 4.3).

**The Researchers.** Site specific apparent indications of anaerobic MTBE biodegradation, based on isotopic fractionation, have been collected and have been published during the last decade by three main authors (J.T. Wilson, T. Kuder and R. Kolhatkar who have worked and published together), and their respective colleagues (see Table 5.3 below)

**The Study Sites.** Much of the data comes from sites in Orange County, California, with one additional site from Parsippany, New Jersey. A common characteristic of these sites is a relatively high concentration of TBA (a co-contaminant of MTBE and a possible degradation product) to MTBE. Data was collected from thirteen sites (each containing multiple monitoring wells) in Orange County during 2002 and 2003. The sites discussed in Kuder et al. (2005) are a subset of the ones discussed in Wilson et al. (2005a). However,

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<sup>22</sup> USEPA 2005 was authored by J.T. Wilson, P.M. Kaiser and C. Adair all with the USEPA Office in Ada, Oklahoma. The document has sometimes been cited as Wilson et al., 2005, which makes things confusing given that Wilson and colleagues had two other publications that year,

while Wilson et al. (2005a) identified each site by the case number assigned to it by the Regional Water Control Board, Kuder et al., (2005) kept the site name and address ambiguous, assigning each the acronym OC (Orange County) followed by a number from 1 to 8 (Table 5.4). This discrepancy gives the mistaken impression that the two papers (*Kuder et al., 2005 and Wilson et al., 2005a*) are unrelated and that there have been many more than 13 sites studied using isotopic fractionation in Orange County. Similarly, data from the site in Parsippany, New Jersey (*USEPA 2005*) was used in more than one paper, by the same group of authors (*Kolhakar et l., 2002; Wilson et al., 2005b, Kuder et al., 2005*), again leading to the impression that the data came from more than one site (Table 5.4).

**Prior and ongoing remediation, and the differential transport properties of MTBE and TBA, were ignored.** While the USEPA sought to establish whether or not MTBE was “naturally” attenuating in groundwater, the USEPA (2005) overlooked the prior and ongoing remediation at the study sites and its potential effects on the isotopic fractionation of MTBE and potential impacts on the relative concentrations of MTBE and TBA (as explained in sections 5.5.1 and 5.5.2 above). In fact, of 12 sites with evidence of MTBE fractionation, 11 sites had either undergone remediation, or were in remediation at the time of sampling (Table 5.4). Groundwater samples were collected from within, or from wells very nearby, remediation wells. The USEPA also ignored the differential fate and transport properties of MTBE and TBA, properties that naturally lead to their fractionation (and changing concentration ratios) in the environment (see Section 5.5.1). These oversights allowed for the mistaken assumption that elevated and/or rising TBA to MTBE ratios, and the isotopic fractionation of MTBE were necessarily the result of natural processes, mainly biodegradation.

**TBA was in the gasoline.** To clarify, TBA is a common impurity in MTBE, accounting for up to 10% of the MTBE added to gasoline (*USEPA 2005; Eweis et al., 2007*). As explained in Section 5.5.1, given the differential fate and transport properties of MTBE and TBA, elevated TBA to MTBE ratios in groundwater are to be expected under natural conditions because these chemicals partition differently between soil, water and air and migrate at different speeds in groundwater. For example, once in groundwater TBA is relatively nonvolatile compared to MTBE. Therefore, remediation processes that include volatilization (evaporation), such as Dual Phase Extraction (DPE), Soil Vapor Extraction (SVE), and Air Sparging (AS), will remove MTBE from groundwater, leaving most of the TBA behind. As discussed in Section 5.5.1, Eweis et al. (2007) showed that volatilization will eventually lead to TBA concentrations that compare favorably to those predicted from the dissolution of gasoline, and MTBE concentrations that are much lower than predicted through dissolution, a signal that could understandably be misinterpreted as MTBE biodegradation.

**Study sites were not under natural conditions.** In light of this background information, Eweis et al. (2007) summarized the research of Wilson et al. (2005a) as follows:

“Wilson et al. [2005a] compared field-measured MTBE and TBA concentrations at contaminated sites in Orange County, California, to those predicted from dissolution of MTBE-oxygenated gasoline into water. Predicted concentrations were based on the assumption that gasoline

contained 11 vol % MTBE, and 0.22 vol % TBA (based on 2% TBA by weight in MTBE). Wilson et al. [2005a] noted that measured TBA concentrations at these sites were in agreement with those predicted from dissolution, whereas the measured MTBE concentrations were commonly one or more orders of magnitude less than predicted from dissolution of MTBE oxygenated gasoline into water. The researchers theorized that “some process or processes” are acting to lower MTBE and increase TBA concentrations [Wilson et al., 2005a]. Using carbon isotope fractionation in a focused study of 13 sites, with TBA/MTBE ratios greater than those expected from the dissolution of fresh MTBE oxygenated gasoline, Wilson et al. [2005a] concluded that MTBE biodegradation could explain the majority of TBA concentrations at a fraction of the sites (4 out of 13), but could not explain the majority of TBA concentrations at a majority of the sites (9 out of 13).”

Wilson et al. (2005a) reported some evidence of biodegradation<sup>23</sup> for 12 of the 13 sites, and no evidence for MTBE biodegradation at one site (at site 99UT015 the two wells tested showed “no evidence of biodegradation” Wilson et al., 2005a).

However, as discussed in Eweis et al. (2007), Wilson et al. (2005a) overlooked the prior and ongoing remediation by volatilization conducted at sites (Table 5.4), as well as a concurrent rise in the water table into the previously unsaturated zone where volatilization had taken place. The latter is important because, as the water table rises, the contamination affected by volatilization in the unsaturated zone enters the saturated groundwater where it can then be detected in the monitoring wells (See Section 5.5.1 and Eweis et al., 2007). Our review of site files from the 13 sites found that (Table 5.4):

- substantial remediation was performed for at least 11 of the 12 sites with reported evidence of biodegradation, and
- no significant remediation was performed at the one site where Wilson et al. (2005a) reported “no evidence of biodegradation”.

In other words, the potential for biodegradation was not studied under natural conditions. Moreover, the isotopic fractionation and elevated and/or rising TBA to MTBE ratios appeared to be associated with the remediation. Not noted by Eweis et al. (2007) was that the remaining site where no reported remediation was performed, and for which Wilson et al. reported evidence of biodegradation, appears to have been associated with a vapor leak.<sup>24</sup>

Rather than present findings from all 13 sites, in Eweis et al. (2007) we chose to present findings from two sites that the USEPA (2005) chose as exemplary of MTBE “biodegradation”. The two figures presented in Eweis et al. (2007) (one in the text and one in the online material of the Journal publication) contain the data presented in USEPA (2005) Figure 5.3 “Transition from MTBE to TBA in monitoring wells at gasoline spill sites in Orange County, California.” The Figure from Eweis et al. (2007) appears in Figure 5.4 presented in the previous section (5.5.1) of this report, discussed earlier in the context of TBA and MTBE volatilization. With regards to the data in the figure, the USEPA (2005) writes:

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<sup>23</sup> Data for isotopic fractionation varied by monitoring well. For example, while one well may have had a strong shift in the isotopic signal, a nearby well had an insignificant shift and was therefore seen as not having exhibited signs of biodegradation.

<sup>24</sup> The site general lacks detectable BTEX.

“The best indication for MTBE biodegradation that is available from conventional monitoring parameters is an abrupt and persistent increase in the ratio of TBA to MTBE in ground water.”

However, as the figures from Eweis et al. (2007) show, the researchers of Wilson et al. (2005a) were sampling sites and specific wells, with concurrent DPE remediation. The additional analysis in Eweis et al. (2007) shows that the observed trends in TBA and MTBE concentrations in those wells are consistent with that expected from the remediation. Accordingly, the most likely cause for the observed changes in TBA to MTBE ratios was clearly the remediation, and not biodegradation<sup>25</sup>.

At the time of the Wilson et al. (2005a) study, the fractionation of stable carbon isotopes (in particular the enrichment of heavy isotopes) detected through CSIA was thought to provide “an unequivocal indication of MTBE biodegradation” (USEPA, 2005). It is noteworthy that similar statements have occurred in numerous publications since the late 1990’s. Specifically, Kolhatkar et al. (2002), Kuder et al. (2005), Wilson et al. (2005a) and USEPA (2005) thought that the isotopic fractionation effects from physical processes were negligible. However, as explained in this report, more recent findings showing that sorption, diffusion and volatilization can all significantly affect isotopic fractionation contradict those notions.

Eweis et al. (2007) called attention to the evolving nature of the research and what we do not yet know about isotopic fractionation. For example, DPE and SVE involve much more than volatilization. Both processes involve the four-phase transfer of MTBE between air, water, soil and free-phase gasoline, as well as the transport of gasoline vapors over tens of feet, and potentially the redistribution of gasoline from a vapor to a free-phase in the vicinity of the extraction well. Isotopic fractionation will occur in all of these mass transfer processes.

As was pointed out in Eweis et al., (2007):

“To our knowledge, the extent to which MTBE isotopically fractionates from these, and other, physical processes has not been assessed at field sites undergoing DPE and/or SVE remediation”.

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<sup>25</sup> 13 Note that similar trends might be observed even if the remediation were not concurrent, but rather occurred earlier in time, because of the regional water level rise into the unsaturated zone above the water table affected by remediation

Table 5.3: Select papers on the anaerobic biodegradation of MTBE:

Citation and year published	Authors	Publication Title	Publishing entity
Kolhatkar et al., 2002	R. Kolhatkar, T. Kuder, P. Philp, J. Allen, and J.T. Wilson	Use of compound-specific stable carbon isotope analyses to demonstrate anaerobic biodegradation of MTBE in groundwater at a gasoline release site	Environmental Science and Technology
Kuder et al., 2005	T. Kuder, J.T. Wilson, P. Kaiser, R. Kolhatkar, P. Philp, and J. Allen	Enrichment of stable carbon and hydrogen isotopes during anaerobic biodegradation of MTBE: microcosm and field evidence	Environmental Science and Technology
Wilson et al., 2005a	J.T. Wilson, R. Kolhatkar, T. Kuder, P. Philp, and S. Daugherty	Stable isotope analysis of MTBE to evaluate the source of TBA in ground water	Ground Water Monitoring and Remediation
Wilson et al., 2005b	J.T. Wilson, C. Adair, P.M. Kaiser, and R. Kolhatkar	Anaerobic biodegradation of MTBE at a gasoline spill site	Ground Water Monitoring and Remediation
USEPA 2005	J.T. Wilson, P.M. Kaiser, and C. Adair,	Monitored Natural Attenuation of MTBE as a Risk Management Option at Leaking Underground Storage Tank Sites	USEPA
USEPA 2007	J.T. Wilson and C Adair	Monitored natural attenuation of tertiary butyl alcohol (TBA) in ground water at gasoline spill sites	USEPA

Table 5.4: Remediation performed at sites used in the natural-attenuation/isotopic-fractionation studies of Wilson et al. (2005a) and Kuder et al. (2005). Thirteen of fourteen sites were in Orange County, California (Page 1 of 2).

Site Name	Address	Wilson et al. (2005b) and USEPA (2005)	Kuder et al. (2005)	Remediation prior to or concurrent with 2002- 2003 Study	Remediation After 2003	Current Site Open/Closure Info
Mobil #18-F1M	5502 Orangethorpe, La Palma, Orange County, California	96UT028	OC1	•SVE •DPE (1999-2000) •DPE (2001-2002)	•Monthly over-purging (2004) •Bioreactor startup (2004) •GWPT (2004-2011).	•OPEN •Begin date 1996
Mobil #18-G3W	15001 Goldenwest, Huntington Beach, Orange County, California	00UT038	OC2	•SE •DPE (2001) •DPE/GWPT system installed (2001) •DPE (2002) •over-purging (2002).	•GWPT system started (2004) •GWE (2008).	• OPEN •Begin Date 2000
Mobil #18-H7Q	13872 Red Hill Ave., Tustin, Orange County, California	88UT138	OC3	•LPH pump outs (1990-1991) •Over-purging (1998-2000) •DPE testing (1999) •Tank upgrades (2000) •150 ton SE (2000) •SVE (2003).	•Over-purging (2006) •Oxygen emitters installed (2007) •Over-purging (2010) •Chemical oxidation injection (2010).	•OPEN •Begin Date 1988
Mobil #18-HL8	26051 La Paz, Mission Viejo, Orange County, California	87UT211	OC4	•Tank upgrades (1987) •SE (1987) •SVE (1991) •DPE (1999-2001)	•167 ton SE (2006) •Fuel system removal activities (2006) •Installed soil vapor intrusion barrier (2008).	•OPEN •Begin Date 1987
Arco #1888	16501 Golden West, Huntington Beach, Orange County, California	88UT198	OC5	•Over-purging (1993) •SVE (1993, 1995) •SVE (2000-2003) •GWPT system (2003)	•Over-purging events (2006-2009) •11 iSOC units (2010-2011)	•OPEN •Begin Date 1988
Arco #1905 <sup>a</sup>	18025 Magnolia, Fountain Valley, Orange County, California	85UT114	OC 6	•SE (1992) •SVE (1992-2000) •Free Product removal (hand bailing; 1997-1999) •DPE (2000-2003)	•DPE (2004-2006) •DPE which consist of separate SVE and GWE systems: •SVE (2007-2010) •GWE (2007-2011)	•OPEN •Begin Date 1985
Shell Oil <sup>b</sup>	16969 Brookhurst, Fountain Valley, Orange County, California	98UT010	OC 7	•No Cleanup actions exist.	•No Cleanup actions exist •Oxygen pulse sparging proposed and six sparge wells installed (2004)	•OPEN •Begin Date 1997
Shell Oil <sup>b</sup>	30011 Crown Valley, Laguna Niguel, Orange County, California	91UT086	OC 8	•Manual Separate-phase hydrocarbons (SPH) recovery (1986-1990) •SVE (1996) •Over-purging (1999) •SE and treat (2003)	•None	•Closed 8/2005 •Begin Date 1991

CONTINUED ON FOLLOWING PAGE

Table 5.4 (CONTINUED): Remediation performed at sites used in the natural-attenuation/isotopic-fractionation studies of Wilson et al. (2005a) and Kuder et al. (2005). Thirteen of fourteen sites were in Orange County, California (Page 2 of 2).

Site Name	Address	Wilson et al. (2005b) and USEPA (2005)	Kuder et al. (2005)	Remediation prior to or concurrent with 2002- 2003 Study	Remediation After 2003	Current Site Open/Closure Info
Shell Oil <sup>b</sup>	27101 Ortega Highway, San Juan Capistrano, Orange County, California	86UT062	-	<ul style="list-style-type: none"> <li>•SVE (1988-1990 and 1992)</li> <li>•GWPT (1989-1996)</li> <li>•DPE (SVE-GWE) (2001)</li> <li>•SE and tank removal (1986, 2000, and 2003)</li> </ul>	<ul style="list-style-type: none"> <li>•None</li> </ul>	<ul style="list-style-type: none"> <li>•Closed 6/2006</li> <li>•Begin Date 1986</li> </ul>
Farjami or Stanton Mobil Station	12493 Beach Blvd., Stanton, Orange County, California	99UT032	-	<ul style="list-style-type: none"> <li>•SE and tank removal (1993)</li> <li>•AS-SVE system (1994-1995)</li> <li>•DPE system (2002)</li> <li>•GWPT (2003).</li> </ul>	<ul style="list-style-type: none"> <li>•GWPT (2004-2008)</li> <li>•Oxygen infusion device (2009-2010)</li> <li>•GWPT (2010-2011).</li> </ul>	<ul style="list-style-type: none"> <li>• OPEN</li> <li>•Begin Date 1997</li> </ul>
Former Texaco Service Station <sup>c</sup>	6011 Manchester Blvd., Buena Park, Orange County, California	86UT175	-	<ul style="list-style-type: none"> <li>•Tank removal (1986)</li> <li>•Free product removal (1987-1988)</li> <li>•GWE and Nutrient enhancement with subsequent AS (1991-1994)</li> <li>•SE and GWE and Free Product removal (1995-1996)</li> <li>•Periodic pump outs using vacuum truck (2002)</li> </ul>	<ul style="list-style-type: none"> <li>•Proposed and denied oxygen injection pilot test (2004)</li> <li>•tank removal (2006)</li> <li>•Over purging (2006).</li> </ul>	<ul style="list-style-type: none"> <li>• Closed 7/2007</li> <li>•Begin Date 1986</li> </ul>
Tesco-76 #7329	34306 Pacific Coast, Dana Point, Orange County, California	99UT015	-	<ul style="list-style-type: none"> <li>•SE (1990 &amp; 1991)</li> <li>•excavation pit dewatering (1992)</li> </ul>	<ul style="list-style-type: none"> <li>•Free Product removal (LNAPL) (2008)</li> <li>•DPE (2008-2009)</li> <li>•132 ton SE (2010)</li> <li>•Onsite and offsite ozone injection and DPE (2010)</li> <li>•monthly LPH removal events (2011)</li> <li>•oxygen injection (2011).</li> </ul>	<ul style="list-style-type: none"> <li>• OPEN</li> <li>•Begin Date 1998</li> </ul>
Unocal #5385 or (76 Service Station No. 255385)	34131 Doheny Park Rd., Dana Point (Also listed under Capistrano Beach), Orange County, California	89UT007	-	<ul style="list-style-type: none"> <li>•SE</li> <li>•Tank, and pipe replacement (1990)</li> <li>•SVE system test (1992)</li> <li>•SVE (1995-1996)</li> <li>•EBD (1995-1996)</li> <li>•ORC (1998)</li> </ul>	<ul style="list-style-type: none"> <li>•AS ozone air sparging system (2004-2009)</li> </ul>	<ul style="list-style-type: none"> <li>• OPEN</li> <li>•Begin Date 1988</li> </ul>
?	Parsippany, NJ	Parsippany	yes	UNKNOWN	UNKNOWN	UNKNOWN

<sup>a</sup>Site used in Busch-Harris et al. (2008) Biotrap study; <sup>b</sup>Site used in Shell DeVaull et al. (2003) study; <sup>c</sup>Site considered in Shell DeVaull et al. (2003) study.

SE: Soil extraction/excavation

SVE: Soil vapor extraction

DPE: Dual phase extraction

AS: Air sparging

GWE: Groundwater extraction

GWPT: Groundwater pump and treat system

iSOC unit: In Situ Submerged Oxygen Curtain

EBD: Enhanced Biodegradation

ORC: Oxygen Releasing Compound (Injected)

### **5.5.2.3 The use of Bio-Traps to verify MTBE biodegradation:**

More recently, <sup>13</sup>C-labeled chemicals, such as MTBE and TBA, have been used to better measure the occurrence of biodegradation *in situ*. In general terms, a bio-trap is made up of a perforated container, packed with activated carbon, which is then allowed to adsorb the chemical in question. When placed in the subsurface (usually for several weeks), the bio-trap acts as a microcosm, where bacteria can grow. If the bacteria degrade the <sup>13</sup>C-labeled chemical, it can incorporate it into its cell mass. CSIA can then be used to analyze cellular material (such as phospholipid fatty acids, or amino acids) to draw a more direct link between the baited chemical and microbial degradation. This method can be useful to show “degradation potential rather than degradation rate” (*Bombach 2010*).

In a study by Busch-Harris et al., (2008), bio-traps were used at one of the original thirteen Orange County sites studied by Wilson et al. (2005a), to examine MTBE and TBA biodegradation *in situ*. The site at 18025 Magnolia street (85UT114; see Table 4.4) had been undergoing remediation, first through SVE (since 1992) and then through DPE starting in 2000. The <sup>13</sup>C-MTBE and <sup>13</sup>C-TBA amended bio-traps were deployed in 2005 and 2006 in wells MW-15 and MW16. Both wells (as well as several others on this site) were used for DPE operation, and remediation in the two wells had to be turned off temporarily while the bio-traps were being incubated in the subsurface. Because of “limited success due to low levels of total biomass collected in the bio-traps” the first experiment with the <sup>13</sup>C-MTBE amended bio-traps had to be repeated using a larger setup, with “five to six times the number of Bio-Sep beads per bio-trap as those used in the first deployment” (*Busch-Harris et al., 2008*). However, very little cellular mass could be extracted from the bio-traps: “no enriched fatty acids were detected in bio-traps from well MW15”, and only one fatty acid was detected at high enough concentration in well MW16. Yet, the authors (*Busch-Harris et al., 2008*) concluded that the results demonstrated the “existence of degradation potential” in the aquifer. However, a similar study with bio-traps (*Fiorenza et al., 2009*) concluded that <sup>13</sup>C-phospholipid fatty acids within the range reported by Busch-Harris et al. (2008) were not considered favorable for natural attenuation through biodegradation.

Of course none of the foregoing details address the salient question: can natural attenuation control the plumes? Some researchers apparently believe that the data from site 85UT114, including isotopic fractionation and elevated TBA to MTBE ratios (*Wilson et al., 2005a and Kuder et al., 2005; Busch-Harris et al., 2008*), stem from biodegradation. As such, site 85UT114 represents one of the more optimistic examples of apparent biodegradation in Orange County, or the country for that matter. Despite this, the plumes are not being controlled, even with substantial remediation efforts. The release was discovered in 1985; remediation, which started at this site in 1992, continues as of the last groundwater monitoring report available on Geotracker (1<sup>st</sup> quarter 2011); and the State Geotracker system describes the “Groundwater Contamination Plume Not Stable or Decreasing”.<sup>26</sup>

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<sup>26</sup> [http://geotracker.swrcb.ca.gov/profile\\_report.asp?global\\_id=T0605900033](http://geotracker.swrcb.ca.gov/profile_report.asp?global_id=T0605900033)

#### **5.5.2.4 Shell Study in Orange County**

In November of 2003, Shell Global Solutions submitted a report to Seth Daugherty of the Orange County Health Care Agency addressing the presence of TBA at selected UST remediation sites in Orange County (*DeVaull et al., 2003*). Shell scientists noted that, depending on the commercial process used to produce MTBE, “TBA may be present in MtBE either as a co-product, an impurity, or as an unreacted initial component, depending on the [MTBE] manufacturing process... Typical gasoline-grade MtBE contains 96 to 99% MTBE by weight” (*DeVaull et al., 2003*). As such, TBA was often present in the MTBE added to gasoline in California. DeVaull et al., (2003) however, could not explain observed concentrations at UST sites (high TBA, low MTBE and low BTEX) simply through physical processes such as “dissolution, partitioning, mixing, and dilution”, and looked for “biological transformation” (of MTBE to TBA) as a potential source for measured high TBA concentrations. Like USEPA (2005), DeVaull et al (2003) did not consider other possible explanations for the elevated TBA to MTBE ratios, e.g., volatilization and remediation (as explained by *Eweis et al., 2007*).

DeVaull et al. (2003) built laboratory microcosms with soil and groundwater samples from three sites in Orange County (see Table 5.4). While six sites were initially considered, only three were sampled for the laboratory biodegradation experiments; those sites were part of the original 13 sites chosen by Wilson et al. (2005a). Biodegradation of MTBE and TBA was examined under different conditions, including aerobic, cometabolic, anaerobic with methanol added (methanogenic) and anaerobic (the latter two being most common at release sites, and therefore more relevant to natural attenuation).

While MTBE and TBA appeared to degrade in many of the microcosms, some of the results seemed to be inconsistent. MTBE and TBA were degraded under aerobic conditions, but under co-metabolic conditions some microcosms from the same site showed degradation while others did not. The authors noted that: “the results from the co-metabolism tests are inconclusive” (*DeVaull et al., 2003*). Under anaerobic conditions (with methanol added to create methanogenic conditions) MTBE was degraded in 8 out of 9 microcosms, while TBA was degraded in 1 out of 9. Finally, under anaerobic conditions (without the addition of methanol), MTBE was degraded in 7 out of 9 microcosms, while TBA was degraded in another 7 out of 9. MTBE and TBA were degraded in the same microcosm in only 5 out of 9 microcosms.

The Shell scientists did not conduct any follow up work to enrich for the microbial cultures, and their work was not published in any scientific journals. To my knowledge, the work by DeVaull et al. (2003) did not undergo an impartial<sup>27</sup> peer review process. This is important considering that some of the laboratory techniques appear suspect or flawed. For example, all of the 18 anaerobic microcosms (designed to be deprived of oxygen) had measurable concentrations of oxygen at the end of the incubation period. In fact, 3 out of 9 microcosms labeled as “anaerobic with methanol” and 6 out of 9 microcosms labeled “anaerobic”

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<sup>27</sup> The report (DeVaull et al., 2003) lists G.E. Spinnler, also a Shell Global Solutions scientist, as the only reviewer.

had oxygen concentrations high enough to qualify as “aerobic,” as defined by DeVaul et al., (2003)<sup>28</sup>.

Furthermore, the results obtained by DeVaul et al., (2003), particularly with regards to TBA degradation under anaerobic conditions, seem inconsistent with the work conducted by the USEPA. In microcosms from six different contaminated sites in the U.S., TBA was degraded under anaerobic conditions in microcosms from only one site (USEPA, 2007). USEPA (2007) noted:

“In summary, the microcosm experiments did not present compelling evidence that anaerobic TBA biodegradation was widespread at gasoline spill sites.”

Furthermore, and based on the literature available at the time, USEPA (2007) noted:

“A close examination of the available information indicates that a default presumption that TBA is readily degraded in anaerobic ground water is not justified”

It should also be noted that DeVaul et al., (2003) were careful not to suggest that the results of their work be extrapolated to other sites in Orange County without further examination:

“Finally, we note that the selected sites exhibited low groundwater gradients and inferred low groundwater seepage velocities. We would expect the effects of biological degradation to be more significant and observable at these types of sites, than at sites with higher groundwater flow rates (high permeability/high recharge sites). This is especially true if degradation in anaerobic conditions is the significant transformation and removal mechanism” (DeVaul et al., 2003).

Interestingly, later publications from Shell Oil scientists seem to emphasize the difficulty in attenuating MTBE and TBA through biodegradation by natural microbial cultures in the subsurface:

- A patent publication by Shell Oil Company scientists for a technology that incorporates patented aerobic microorganisms and granular activated carbon for above ground water treatment notes:

“The use of bacteria or naturally occurring microbes for biodegradation of a wide range of organic contaminants is known. However, attempts to degrade MTBE and/or TBA using bacterial cultures have generally met with little success” (Byers et al., 2004).

- Also in a patent publication by Shell Oil Company scientists for a technology that involves introducing patented aerobic microbial cultures and oxygen into the subsurface for the treatment of “ethers and/or alcohol” the scientists noted:

“Now, having used oxygenated fuels for several years, it has become clear that these cleaner-burning fuels pose great threats to groundwater resources. In particular, many oxygenate chemicals are very soluble in water and are slow to degrade in the environment; hence they tend to accumulate in water resources once released to the environment” (Salanitro et al., 2005).

The scientists continued to state:

“It is also now known that when oxygenate chemicals including alkyl ethers, such as MTBE and tertiary butyl alcohol [TBA], are found in the subsurface, then they are resistant to biodegradation under natural conditions. This is the main reason for their persistence and accumulation in soil and groundwater” (Salanitro et al., 2005).

As such, the USEPA (2007) document on the natural attenuation of TBA states:

“If a transport and fate model is used as part of the risk evaluation, there is no justification to include biodegradation in the model. Including biodegradation in the model requires direct

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<sup>28</sup> DeVaul et al., (2003) noted: “aerobic criteria for dissolved oxygen > 1 mg/L”

evidence that bacteria in the contaminated aquifer are capable of degrading TBA, or in fact, have degraded TBA” (USEPA 2007)

Finally, it is important to note that several of the sites originally considered for the study by DeVaul et al. (2003) continue to undergo engineered remediation and were open as of the last monitoring period reported on Geotracker in 2011.

## 5.6 Migration of MTBE to Supply Wells

Contamination entering groundwater can travel to water supply wells. Because contamination moves with the groundwater, travel times from contaminant source areas to supply wells commonly range from years to centuries, depending on hydrologic conditions and pumping rate. Because supply wells commonly pump a mix of water from various recharge locations, contamination entering from any particular release location will tend to mix with uncontaminated water from other locations, diluting detectable concentrations in the well (*Einarson and Mackay, 2001*). This process is called wellbore dilution.

An MTBE plume may be diluted in a well due to wellbore mixing (*Johnson et al., 2000*). Because public-supply wells typically have long screened intervals, wellbore dilution is generally appreciable.

Consequently, low-level MTBE contamination from non-point sources, such as atmospheric deposition, is unlikely to result in detectable (with standard methods) levels of contamination in an operating supply well. This is an important point because atmospheric sources of MTBE contamination have been shown to produce surface water or shallow groundwater MTBE concentrations of about 1 µg/L: “maximum concentrations of MTBE that might occur in shallow groundwater as a result of atmospheric deposition would be approximately 1 µg/L” (*NJDEP, 2001*). Thus, the detection of MTBE concentrations in public water supply wells generally implicates a contaminant source having a much higher concentration than that of a nonpoint source. The mere detection of MTBE in a public supply well strongly implicates point sources, such as LUFT sites, of contamination. These concepts are illustrated in Figure 5.6a and 5.6b. Importantly, if the source is a LUFT and the core of the plume emanating from the LUFT is en route to the well, then continued pumping of the contaminated water supply well will typically result in ever increasing MTBE concentrations in drinking water produced by the well.

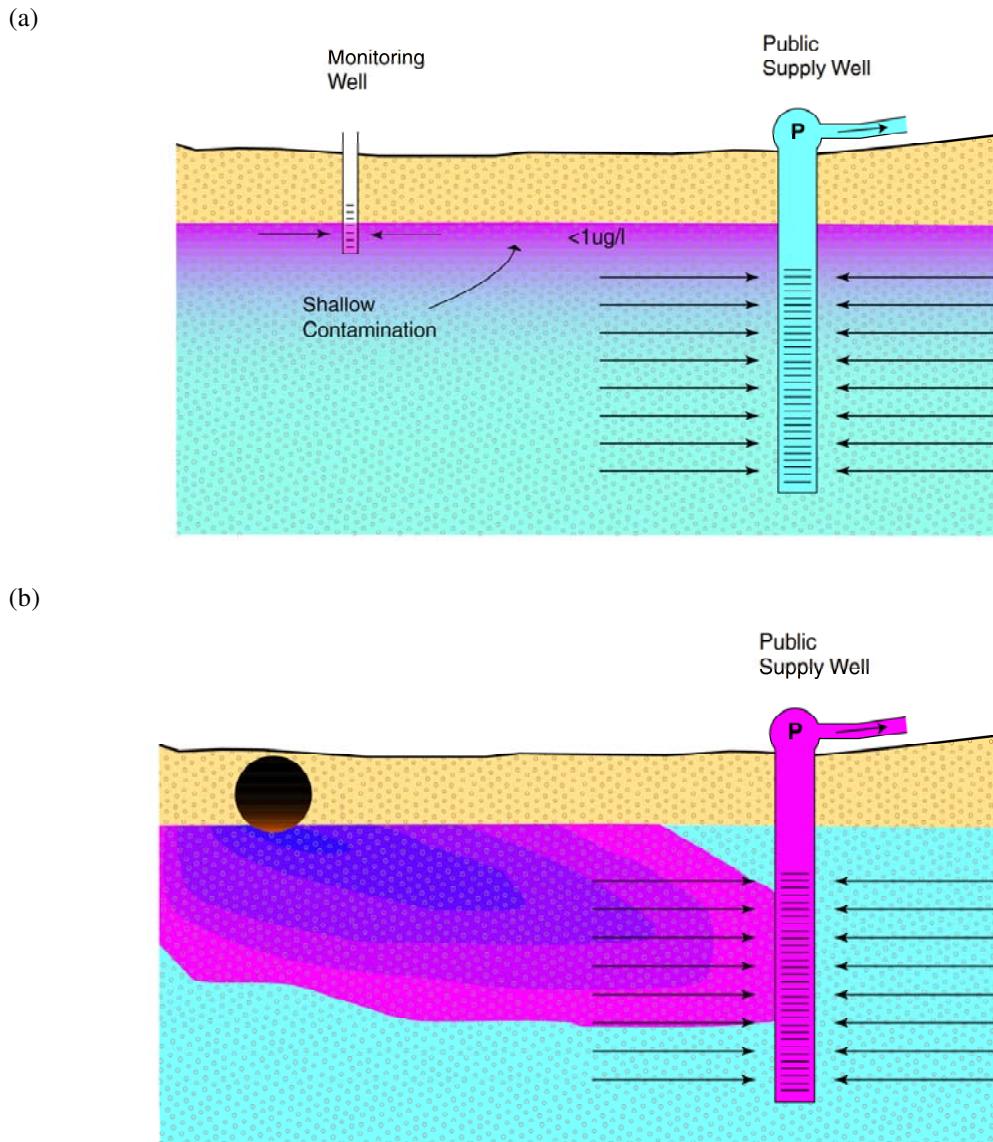


Figure 5.6. (a) Schematic representation of low-concentration, diffuse source of MTBE to a public water supply well. Dilution with uncontaminated groundwater entering the well will commonly result in the measured MTBE concentration at very low values, or below detection. (b) Schematic representation of a high-concentration, point-source of MTBE, which is likely to produce measurable or elevated concentrations of MTBE in public water supply well water samples.

## 5.7 The Role of Time

“[Groundwater] velocities under typical hydraulic gradients can range from a few millimeters per year to a meter per day” (*Squillace et al., 1998*). As such, travel times from recharge locations to discharge locations, such as an individual supply well, commonly range from years to centuries (e.g., *Fogg et al., 1998a; Johnson et al., 2000*). The time a ‘packet’ of water spends in the groundwater is referred to as its age. Although groundwater ages are commonly expressed as average values, water pumped from a well is derived from a mix of different recharge locations resulting in a mix of different ages (times of travel to the well) in any given sample (e.g., *Busenberg and Plummer, 1992; Fogg et al., 1998a; Weissmann et al., 2002; Bethke and Johnson, 2002*). Additionally, supply wells will commonly draw a significant fraction of older water (*Fogg et al., 1998a; Weissmann et al., 2002*). Therefore, it would not be surprising, for example, to find groundwater with an average age of 50 years resulting from a mix of waters that range from tens to hundreds of years in age.

### 5.7.1 Volatile Organic Compounds

The USGS studied the occurrence of 86 volatile organic compounds (VOCs) in groundwater to assess the vulnerability of particular supply wells in the Los Angeles, California basin (*Shelton et al., 2001*). The basin is recharged primarily from engineered facilities. Of the 86 VOCs, the study found MTBE to have the most localized distribution (primarily near recharge facilities). The authors concluded that: “this nearly exclusive occurrence of MTBE close to the area of recharge is consistent with the relatively recent introduction of MTBE as a fuel additive.” In contrast, chloroform contamination introduced in the early 1900’s was detected more widely and at greater distances from the recharge facilities than all other VOCs. Similarly, a vulnerability assessment of California wells in the Livermore-Amador and Niles Cone groundwater basins found “MTBE is a good indicator of very recently recharged water.”

Because MTBE is generally associated with “recently recharged [young] water,” not enough time has elapsed for many MTBE plumes to spread to drinking water wells. Travel times from contaminant source locations to wells can range from days to a century or more, with times on the order of decades quite common in many basins (*Fogg, et al., 1998a; Weissmann et al., 2002*). The recent introduction of MTBE relative to other persistent groundwater contaminants can be seen in Table 5.3 below.

Table 5.3. Historical Use of Selected Anthropogenic Groundwater Contaminants in California.

Compound	Principal Uses and Source(s) to Groundwater	Widespread Use in US	MCL ( $\mu\text{g/l}$ ) <sup>e</sup>
Nitrate	Agricultural fertilizer, feed lots	~1950 – present	45,000
TCE	Industrial solvent	~1930 – present <sup>d</sup>	5.0
PCE	Industrial solvent	~1930 – present <sup>d</sup>	5.0
DBCP	Agricultural soil fumigant /nematocide <sup>a</sup> (banned in 1977)	~1960 – 1977 <sup>a,b,c</sup>	0.2
MTBE	Gasoline LUFTs	1992-2006	5.0 <sup>f</sup>

a: <http://www.epa.gov/ttn/atw/hlthef/dibromo-.html>, Accessed March 24, 2004

b: Raski, D. J., I. J. Thomason, J J. Chitambar, and H. Ferris, September, 2003, A History of Nematology in California, available at <http://plpnemweb.ucdavis.edu/nemaplex/History%20of%20Nematology%20in%20California.pdf>

c: Burow et al. (1999)

d: <http://www.concentric.net/~Rnk0228/solhist.html>

e: <http://www.epa.gov/safewater/hfacts.html> - Synthetic

f: Secondary taste and odor MCL for California.

### 5.7.2 Vulnerability to Contamination

The vulnerability of a water supply well to contamination depends upon the travel time from contaminant source areas to the well screen or open hole in bedrock, which in turn, depends in part upon hydrologic conditions, well placement, well construction, and pumping rate. As a general rule, shallow groundwater is more vulnerable to contamination than deep groundwater, due to its proximity to the land surface, and therefore contaminant source areas. Private wells are generally more susceptible to contamination than public wells, as they tend to be shallower and of poorer construction with regards to minimizing contamination (*Happel et al.*, 1998). Drinking water wells in New Jersey are highly vulnerable to contamination as evidenced by several of the sites at issue in this case.

The advent of widespread use of MTBE in gasoline is relatively recent, and therefore many MTBE plumes are still young. By comparison, many other contaminants that are less soluble (Figure 5.1) and less mobile than MTBE, for example solvents such as perchloroethylene (PCE) and trichloroethylene (TCE), have been in widespread use for many decades (*Shelton et al.*, 2001; Table 5.3). Contaminants like these, released into groundwater in decades past, are commonly more widespread than MTBE in older, deeper groundwater.

Their observed fate alerts us to the likely long-term fate of MTBE.

## 6 Remediation and Natural Attenuation of MTBE in Groundwater

### 6.1 Remediation

Persistent contamination can render groundwater unusable for decades to centuries, absent costly and time-consuming remediation or expensive treatment. “Once a groundwater supply is polluted, it is [often] difficult and expensive to clean up” (*Helpern et al., 2001*). This is largely due to the same groundwater flow and transport processes that cause significant variations in groundwater age within the same aquifer system. That is, once an aquifer is exposed to a dissolved contaminant like MTBE, much of the contaminant migrates into regions of extremely slow groundwater flow (e.g., silts and clays). From there, the contaminants can migrate, or “bleed,” out very slowly from the slow-flow regions over an extended period of time (*LaBolle and Fogg, 2001; Zhang et al., 2007*). A phenomena, often called “tailing,” describes a situation in which the rate of bleed-out is slow enough to allow the contaminant to persist at problematic concentrations for decades to centuries. This “tailing” is the primary reason that decontamination of aquifers can take decades to centuries longer than contamination of aquifers (*LaBolle and Fogg, 2001; Zhang et al., 2007*).

### 6.2 Natural Attenuation

Natural attenuation is the term used to describe contaminant removal from the environment (typically the subsurface) through natural processes, without engineered intervention. These natural processes can be physical, chemical, or biological. Where applicable, the use of monitored natural attenuation as a remediation strategy can reduce the cost of site cleanup by millions of dollars.

The increased reliance on natural attenuation for cleanup of contaminated groundwater in the 1990’s, prompted the release of “Natural Attenuation for Groundwater Remediation” by the National Research Council (*NRC, 2000*). The objective of the study, which was conducted by 15 experts in the fields of science, technology, and policy of natural attenuation, was to provide guidance on natural attenuation and when and how it should be applied to remediate contaminated soil and groundwater (*Rittmann, 2003*). Natural attenuation was deemed acceptable when the processes involved resulted in the destruction or immobilization of the contaminant (in this context, processes such as dilution, dispersion, and partial breakdown of a contaminant to toxic byproducts, would be considered unacceptable). The Environmental Engineering Committee (EEC) of the USEPA Science Advisory Board (SAB) (*USEPA, 2001*) reviewed the findings of NRC (2000). The committee (SAB) endorsed the findings of NRC 2000, stating:

“During the course of this review, the Subcommittee [Natural Attenuation Research Subcommittee] agreed with and endorsed the overall strategy that NRC (2000) provides for deciding whether or not MNA protects human health and the environment. According to NRC

(2000), MNA should be selected only when a **cause-and-effect relationship is established between loss of contaminant and destruction or immobilization processes.**" (*USEPA 2001*, emphasis added).

The report (*USEPA, 2001*) further stated:

"the Subcommittee considers MNA to be a remediation approach based on understanding and quantitatively documenting naturally occurring processes at a contaminated site that protect humans and ecological receptors from unacceptable risks of exposure to hazardous contaminants. The Subcommittee stresses that, MNA is a "knowledge-based" remedy because, instead of imposing active controls, as in engineered remedies, scientific and engineering knowledge is used to understand and document **naturally** occurring processes." (emphasis as published).

Given that MTBE is not strongly adsorbed to soil matter, *NRC (2000)* listed biotransformation, or biodegradation, as the "dominant attenuation process" for MTBE. *USEPA 2001* explains:

"The NRC (2000) report *de facto* restricted the acceptable (or eligible) natural attenuation processes to biodegradation and strong immobilization. This is a more restrictive definition of that used by EPA...and reflects NRC's finding that, while other processes occur and can contribute to natural attenuation, the processes that destroy or strongly immobilize contaminants are more reliable in reducing risks." (emphasis as published).

In evaluating MTBE and the potential for natural attenuation as a remediation strategy, *NRC (2000)* referred to MTBE as "generally resistant to biodegradation". *NRC (2000)* also rated the likelihood that natural attenuation will succeed as a remediation strategy for MTBE as low. In contrast, the BTEX group was given a rating of high.

In light of more recently published literature on MTBE biodegradation, *Rittmann (2003)*, chairman of the committee responsible for *NRC (2000)*, conducted a reevaluation of the *NRC (2000)* guidance on MTBE natural attenuation. In this update, *Rittmann (2003)* concluded:

"Based on laboratory and field experience so far, the most promising type of MTBE biodegradation is aerobic and involves initial monooxygenase reactions. The situation requires that the supply of dissolved oxygen be significant and that MTBE degraders expressing the monooxygenase enzymes be present in significant numbers. Having both requirements present is not a high probability circumstance for natural attenuation; therefore, the original *NRC (2000)* judgment that the likelihood of success is low for natural attenuation of MTBE seems to remain valid."

Intrinsic bioremediation is described by the National Research Council (*NRC, 1994*) as "the innate capabilities of naturally occurring microbes to degrade contaminants without taking any engineering steps to enhance the process." Evidence collected in the scientific literature thus far suggests that MTBE lacks the "intrinsic bioremediation" ability that naturally regulates BTEX releases. Three examples from oil industry (Shell Global Solutions) studies are given below:

(1) *Salanitro et al. (1999)* state:

"MTBE, the common gasoline oxygenate that has been detected in groundwater, is inherently difficult to biodegrade in aquifer soils because of its ether bond and tertiary carbon features.

Microbes grow slowly and produce low cell yields when cultured on MTBE. Characterization of plumes and aquifer material from many sites indicate that the transport of ether may extend beyond BTEX plumes. The low concentration of indigenous degraders, presence of low dissolved oxygen and the poor growth of microbes on MTBE indicate that natural bioattenuation will be difficult to demonstrate in groundwater plumes.”

(2) Similarly, *Spinnler et al. (2001b)* have concluded:

“So far, the evidence indicating intrinsic bioremediation of MTBE is inconclusive. In fact, the extreme length of some MTBE plumes implies intrinsic bioremediation does not occur at rates capable of controlling plume migration. … Microcosm experiments done in our laboratory from soil collected at MTBE contaminated sites indicated MTBE-degrading microorganisms were present at few sites. The presence of natural degraders does not, by itself, insure biodegradation will be successful. Degraders need to be present in sufficient quantities and respond to stimulation.”

(3) *Spinnler et al. (2001a)* also stated:

“MTBE degraders have not been observed to be as ubiquitous in the environment as BTEX degraders. Even when present, MTBE degraders may not have sufficient population density to degrade high concentrations of MTBE. Laboratory data indicates MTBE is an extremely poor growth substrate for the organisms, so rapid population growth may not occur in contaminated aquifers.”

These observations on the potential for MTBE attenuation through biodegradation under natural conditions have not changed much over the years: For example, based on a review of the literature on MTBE and TBA biodegradation in the subsurface, *Schmidt et al. (2004)* concluded:

“In general, compared with hydrocarbons present in gasoline, fuel oxygenates biodegrade much slower, if at all. The presence of MTBE and related compounds in groundwater therefore frequently limits the use of in situ biodegradation as remediation option at gasoline-contaminated sites. Though degradation of MTBE and TBA in field studies has been reported under oxic conditions, there is hardly any evidence of substantial degradation in the absence of oxygen.”

*Schmidt et al. (2004)* also added: “Reliable in situ degradation rates for MTBE and TBA under various geochemical conditions are not yet available”.

Similarly, and in reference to assessing degradation rates for field conditions, the USEPA document on the natural attenuation of MTBE (*USEPA, 2005*) cautions:

“Most studies in support of MNA do not provide a solid estimate of the rate of degradation of the contaminant in the ground water along the flow path. Without a solid estimate of the rate of degradation in the ground water, a conservative evaluation of the risk to a receptor is restricted to the assumption that the contaminant does not degrade at all.”

Further, *USEPA-OGWDW (2008)* noted:

“MTBE has several properties that increase its persistence and mobility in the environment once released” and “Once dissolved in ground water, it generally resists degradation and moves at nearly the same velocity as ground water”.

At the same time, *Youngster et al. (2008)* stated:

“The prevalence of anoxic conditions in gasoline-polluted groundwater means that anaerobic MTBE degradation will likely be a necessary process of remediation. MTBE degradation is a rare process and the responsible organisms are sparsely distributed, even in contaminated sediments.”

**Natural Attenuation and Transport, MTBE compared to BTEX:** The fact that MTBE is poorly attenuated in the unsaturated zone (the zone between the land surface and the water table) may cause it to be the only contaminant detected in groundwater after a gasoline spill, as was observed in one case in Maine (*Garrett et al., 1986*). Similar observations were noted during a field study in Wisconsin (*McLinn and Rehm, 1997*) designed to evaluate the fate of residual hydrocarbons in the vadose zone. During 2 years of monitoring, MTBE was the only gasoline constituent detected in the groundwater. In the same time period, pore gas samples showed elevated carbon dioxide concentrations and low oxygen concentrations, suggesting that the petroleum hydrocarbons were being degraded in the unsaturated zone above the water table, before making it to the groundwater.

Similarly, because of its transport properties and poor natural attenuation, MTBE plumes tend to travel farther and deeper than BTEX plumes (Table 6.1). In a survey of state experiences with MTBE and other oxygenate contamination at LUST sites (*NEIWPCC, 2003*) 32 states indicated that MTBE plumes tend to be longer than typical BTEX plumes, yet only 11 states require three-dimensional characterization of the plumes. Less than half of the states said that they are taking extra steps to ensure that oxygenates do not migrate beyond standard monitoring parameters.

**Table 6.1: Summary of field observations comparing MTBE plumes to those of BTEX**

Author	Sites Studied	Findings
Dernbach, 2000	Tahoe Basin	MTBE plumes spread at least 3 to 6 times the distance of the largest BTEX plumes.
Salanitro et al., 2000	Port Hueneme, California	MTBE plume traveled more than 1,500m down gradient from the spill source, compared to just 50m for the BTEX components.
Thuma et al., 2001	Deer Park, New York	MTBE plume extended to about 8000 ft, compared to approximately 700 ft for BTEX.
Wilson et al., 2002	Vandenberg Air Force Base in Central California	MTBE plume traveled 17 to 34 times the distance that the BTEX plume did (520 meters for MTBE compared to 15-30 meters for BTEX).
Martienssen et al., 2006	Leuna, East Germany	MTBE plume about 1700 m long and several hundred meter wide. Benzene (longest of the BTEX) plume less than 300 m long.

These observations are consistent with the scientific understanding of MTBE and BTEX transport in groundwater (*Johnson et al., 2000*). For example, in the Tahoe basin, *Dernbach (2000)* noted that the MTBE plumes spread at least 3 to 6 times the distance of the largest BTEX plumes. In comparison, the MTBE plume at Vandenberg Air Force Base in Central California traveled 17 to 34 times the distance that the BTEX plume did (*Wilson et al., 2002*). At the Port Hueneme site, California, the MTBE plume traveled more than 1,500 meters (4,920ft) down gradient from the spill source, compared to just 50 meters (164ft) for the BTEX components (*Salanitro et al., 2000*). In Leuna, East Germany, a MTBE plume extends about 1700 m (5,577ft) in length and several hundred meters wide. The corresponding benzene plume (longest of the BTEX compounds) is less than 300 m (984ft) long. In comparison, BTEX plumes in California rarely exceed 250 feet (76 meters) (*Rice et al., 1995*). In 1995, MTBE in the then nearly 7,000 ft long plume in East Patchogue, N.Y. had already traveled approximately 1,000 ft farther than BTEX (*Weaver et al., 1996*). In fact, because of its persistence and mobility in the subsurface, MTBE has been relied upon as a “conservative tracer” to study the fate of BTEX constituents in the subsurface (*Robbins and Gilbert, 2000; Rifai et al., 1995*), and to conduct forensic investigation of gasoline spills, including the estimation of the timing or source of a gasoline spill (*Davidson and Creek, 2000*).

**MTBE, TBA and Natural Attenuation:** Another potential problem with MTBE biodegradation is the formation of byproducts, particularly TBA, a chemical that is more toxic than MTBE (*Keller et al., 2000*). TBA has been shown to accumulate, before being degraded at a rate slower than that of MTBE (*Salanitro et al., 1994; Bradley et al., 2001a; Wilson et al., 2002*). In other instances, TBA persists or degrades very slowly (*Mormile et al., 1994; Hardison et al., 1997; Hyman et al., 1998; Vainberg et al., 2006; Skinner et al., 2008*). However, although TBA is sometimes observed to be a byproduct of MTBE biodegradation, its mere presence should not be considered proof of natural attenuation or biodegradation of MTBE. Moreover, as discussed previously, elevation in the TBA/MTBE ratio is not proof of natural attenuation or biodegradation of MTBE either.

### 6.2.1 Site characterization and natural attenuation

Many MTBE contamination sites are poorly characterized. One problem stems from the fact that monitoring networks were often developed with BTEX contamination in mind. However, MTBE and BTEX plumes often require different levels of monitoring. The American Petroleum Institute (*API, 2000*) explained:

“If a BTEX release at a given site predates an ether oxygenate [MTBE] release, then the BTEX plume may be as long (or even longer) than the oxygenate plume. However, the older BTEX plume is more likely to have been affected by changing flow directions or fluctuating water levels, while the younger oxygenate plume is more likely to have experienced fewer variations in groundwater conditions. Given these factors, the older BTEX plume may be wider and thicker, and the younger oxygenate [MTBE] plume may be narrower and thinner. Thus, an ether oxygenate [MTBE] plume may be more difficult to detect and delineate, even when its overall length is similar to that of the associated BTEX plume.”

This characteristic of MTBE plumes has been noted by other authors as well:

“MTBE plumes are often narrow compared to BTEX plumes. Unless monitoring well networks are spaced fairly closely across the width of the plume, it is possible to miss the MTBE plume entirely as it slips through between monitoring points. During site characterization, closely spaced direct-push sampling points at multiple depths placed at right angles to the long axis of the plume can be used as a tool for locating a narrow plume and facilitating proper placement of permanent monitoring wells.” (*Ellis, 2000*)

In addition, Fiorenza and Rifai (*2003*) noted that:

**“One of the challenges in assessing natural attenuation at MTBE-contaminated sites has been due to the limitations of the site characterization. For example, the monitoring well network at a site may not be extensive enough, and it might lack depth-discrete sampling intervals that would allow for the characterizations of MTBE and co-contaminants.”**

Often, MTBE plumes may be so large that they have migrated offsite. Existing monitoring networks would be inadequate to detect them as they spread. In fact, *Mace et al.* (*1997*) in their survey of more than 600 gasoline contamination sites concluded that:

**“almost 40 percent of the [600 plus] sites [surveyed] have dissolved hydrocarbon plumes that reportedly extend offsite” (*Mace et al., 1997*)**

Note that the survey was conducted in the 1990s and already MTBE plumes had migrated offsite.

Similarly, in a study of 127 sites by Rifai and Rixey (*2004*) (also discussed in *Rifai et al., 2003*):

**“The MTBE and benzene plumes at many of the sites had traveled off-site and the existing monitoring networks did not capture the full extent of the plume. During the course of the research, it was determined that the monitoring well network at 91 sites [out of 127 examined] was not sufficient for delineating the dissolved phase hydrocarbon plumes; in almost every case this was true for both MTBE and benzene plumes.”**

Clearly monitoring was inadequate.

Quite often, monitoring of MTBE plumes does not account for the vertical migration of contamination.

Very often, monitoring is limited to shallow depths and does not account for vertical contaminant migration deeper into the aquifer. For example, deficiencies in plume monitoring were highlighted in a study of a gasoline release in East Patchogue, New York, that resulted in a 5000 feet long benzene plume and a similarly sized MTBE plume (due to its more recent release), where researchers concluded that:

"If the service station LUST site had been characterized in a traditional manner, without vertical delineation, the estimated plume length and detachment of the MTBE plume would have been completely misinterpreted; the benzene plume at the site would have been judged to be 3,500 feet shorter than actual and the MTBE pulse would have been mischaracterized as below the State of New York's level of concern. [detected MTBE concentrations were as high as 9,600 ppb]" (*Delaware Department of Natural Resources and Environmental Control, August, 2000*).

For all these reasons, Shih et al. (2004) stated:

"Clearly, plume length as defined is two-dimensional. The lack of depth-specific data and other site-specific knowledge across the population of LUFT sites investigated in this paper preclude evaluation of plume transport in the vertical direction. In areas of significant recharge, this can bias the measurements toward shorter plumes, since a typical monitoring well screened across the water table may fail to detect the leading edge of the plume as it is deflected downward in response to the infiltration of recharge from above. Further, fluctuating flow directions as well as errors in their determination can result in monitoring well network configurations that create additional biases in plume length measurement."

Similarly, USEPA (2005):

"Conventional monitoring wells can provide an incomplete picture of the true distribution of MTBE in ground water. If the screen of a monitoring well is long compared to the thickness of the plume of contamination, it can sample the plume of contaminated ground water and cleaner ground water above or below the plume, giving a false impression of natural attenuation from one well to another. Long plumes of MTBE may dive below the screens of monitoring wells altogether. Any evaluation of natural attenuation between monitoring wells should consider the screened intervals of the wells, the depth interval contaminated with gasoline (if that information is available), and the lithological features sampled by the wells."

In addition, MTBE plumes can migrate laterally in perched groundwater above the water table. In such cases, a plume could migrate offsite before it even reaches the water table. Wells drilled to saturated groundwater on site could miss the plume entirely.

### **6.2.2 Lessons Learned from Other Recalcitrant Groundwater Contaminants**

The fact that MTBE has been shown to biodegrade in some studies does not justify reliance on natural attenuation as a contaminant regulation mechanism in the environment. For perspective, contaminants such as DBCP, nitrate, PCE, TCE, and TCP also have been known to biodegrade under a variety of conditions; yet, as discussed previously, concentrations of these compounds have increased in deeper groundwater on a decadal time scale (*Fogg et al., 1998a; Dubrovsky et al., 1998*). Some 30 years since it was banned from use and some 45 – 50 years after its first

widespread use in California, DBCP<sup>29</sup>, known to slowly biodegrade, is still one of the most commonly detected contaminants in PWS wells at concentrations above its MCL (*CDHS database, March, 2006*). The mere existence of conditions under which a compound might biodegrade neither guarantees, nor implies, that the contamination will generally be eliminated or reduced significantly through biodegradation.

## 7 Use of MTBE and other Oxygenates in Gasoline

The following information on the use of MTBE and other common oxygenates [ethyl tertiary butyl ether (ETBE), tertiary amyl methyl ether (TAME) and ethanol] as gasoline fuel additives in the United States comes from information available through the U.S. Energy Information Administration (EIA)<sup>30</sup>:

Oxygenates were first required in 39 areas of the country with the 1992 wintertime oxygenated gasoline program, arising from the Clean Air Act and designed to reduce emissions of carbon monoxide. This program required oxygen at a minimum level of 2.7 percent by weight (which by volume is equivalent to 15.0 percent MTBE or 7.4 percent ethanol). California obtained a waiver from this minimum 2.7 percent oxygen level because of concerns it would increase automotive emissions of oxides of nitrogen (NO<sub>x</sub>). California's requirement was amended to require between 1.8 and 2.2 percent by weight oxygen. The reformulated gasoline program, aimed at reducing emissions of ozone-forming VOCs, nitrogen oxides, and other air pollutants, was implemented year round in certain areas of the United States, and during the summer months in other parts, beginning in December 1994. The program required a minimum of 2.1 percent oxygen by weight (which by volume is equivalent to approximately 11.7 percent MTBE or 5.8 percent ethanol).

MTBE usage as an octane booster grew in the early 1980's in response to the phasing out of lead from gasoline, and an increase in demand for gasoline. The petroleum industry responded to the oxygenated and reformulated gasoline programs by amending gasoline with various oxygenates, principally MTBE and ethanol (Table 7.1). The use of MTBE increased significantly from about 83,000 in 1990 to about 161,000 barrels per day in 1994. MTBE use in gasoline increased to an estimated 256,000 barrels per day by 1997, following the reformulated gasoline program in December 1994 (Table 7.1).

In 2005, the New Jersey Legislature passed legislation that would ban the sale of gasoline containing more than 0.5% MTBE in the State, effective on January 1, 2009.<sup>31</sup>

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<sup>29</sup> Reported half lives for conditions similar to in-situ groundwater range from 6.1 to 141 years (Burow et al., 1999). Also see US EPA Technical Fact Sheet on Dibromochloropropane, DBCP, available at: <http://www.epa.gov/OGWDW/dwh/t-soc/dbcp.html>

<sup>30</sup> <http://www.eia.doe.gov/emeu/steo/pub/special/mtbe.html#Who%20gets%20gasoline%20with%20oxygenates> last accessed 2/15/07

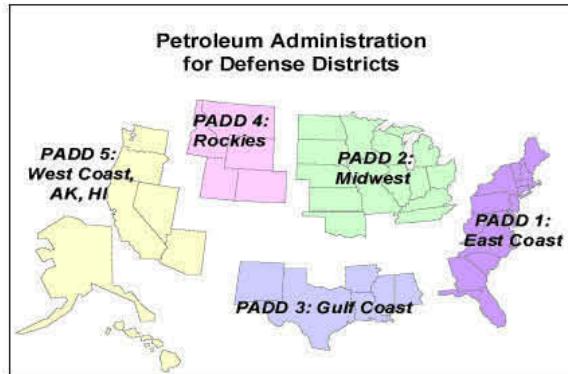
<sup>31</sup> New Jersey Department of Environmental Protection <http://www.state.nj.us/dep/dsr/mtbe-report.htm> last accessed Oct. 26, 2012.

**Table 7.1: Estimated Oxygenate Use in 1997 by Petroleum Administration Defense District (PADD) (expressed in thousand barrels per calendar day).**

	PADD 1	PADD 2	PADD 3	PADD 4	PADD 5	Total	
<b>Reformulated</b>	MTBE	109.80	4.20	25.81	-	97.23	237.03
	ETBE	0.00	0.00	0.00	-	0.34	0.34
	TAME	8.66	0.02	2.97	-	2.94	14.58
	Ethanol	0.74	22.54	0.00	-	0.95	24.22
<b>Oxygenated</b>	MTBE	-	0.00	0.00	0.34	0.46	0.80
	ETBE	-	0.00	0.00	0.00	0.00	0.00
	TAME	-	0.00	0.00	1.08	0.00	1.08
	Ethanol	-	8.96	1.72	2.48	5.14	18.29
	<b>MTRF</b>	<b>17.83</b>	-	-	-	<b>0.10</b>	<b>17.93</b>

## Petroleum Administration for Defense Districts

Use your browser's "back" button to return to where you were,  
or click here to return to [Table of Contents](#)



PADD's were delineated during World War II to facilitate oil allocation.

## 8 MTBE in Groundwater Resources

Due to its widespread use, MTBE is now “... ubiquitous and can be detected at very low concentrations nearly anywhere on the Earth’s surface (*Science and Technology Review, May 2003*).” Widespread detections of MTBE in groundwater resources were seen across the nation (*Report by the Blue Ribbon Panel on Oxygenates in Gasoline, 1999*). In the Executive Summary and Recommendations section of the *Report by the Blue Ribbon Panel on Oxygenates in Gasoline (July 1999)*, the authors state:

“At the same time, the use of MTBE in the program [the Federal Reformulated Gasoline Program] has resulted in growing detections of MTBE in drinking water, with between 5% and 10% of drinking water supplies in high oxygenate use areas showing at least detectable amounts of MTBE. The great majority of these detections to date have been well below levels of public health concern, with approximately one percent rising to levels above 20 ppb [parts per billion which is the same as µg/l]. Detections at lower levels have, however, raised consumer taste and odor concerns that have caused water suppliers to stop using some water supplies and to incur costs of treatment and remediation. The contaminated wells include private wells that are less well protected than public drinking water supplies and not monitored for chemical contamination.”

### 8.1 The Onset of MTBE Contamination in New Jersey

One of the first published reports of MTBE contaminating a community water supply well comes from the Rockaway Township in north-central New Jersey. The town of 20,000 people relied primarily on three wells drilled into unconsolidated glacial deposits at depths of 100 to 200 ft (McKinnon and Dyksen, 1984). In November 1979, TCE was discovered in two of the wells, which resulted in those wells being taken out of service. The remaining well, the largest of the three (pumping at 1,000 gpm) “was not affected and remained in service” (McKinnon and Dyksen, 1984). However, in October 1980, the fuel oxygenates, diisopropyl ether (DIPE) and MTBE “were found at sufficiently high levels to cause severe taste and odor problems in the water...As a result, numerous taste and odor complaints were received from the township’s residents” (McKinnon and Dyksen, 1984).

To treat the groundwater, the township decided to use granular activated carbon (GAC). Based on the TCE levels detected in the wells initially, the GAC beds were projected to last six to eight months before it was necessary regenerate the beds. However, “after three months of operation, the concentration of DIPE in the contactor effluent was 14 µg/L, MTBE was 23 µg/L and TCE was not detected. Therefore, it was necessary to replace the carbon every two months at a cost of about \$32,000. By the end of 1981, the carbon was being replaced at a rate of about once every four to six weeks” (McKinnon and Dyksen, 1984). As a result, an aeration system was added to the treatment train, to help strip the dissolved contaminants from the water before it was passed through the GAC beds. This helped cut the cost of treatment significantly as it added to the lifetime of the GAC beds. However, this case highlighted the difficulty in treating MTBE contaminated drinking water.

## 8.2. USGS studies

The United States Geologic Survey (USGS) has conducted numerous studies on water quality across the nation. Several of those studies addressed contamination in the Eastern United States, and in New Jersey in particular. In many of those studies MTBE was found to be among the most frequently detected volatile organic compounds (VOCs) as explained below.

- **“Occurrence of nitrate, pesticides, and volatile organic compounds in the Kirkwood-Cohansey aquifer system, southern New Jersey”** (*Stackelberg et al., 1997*). This was one of the earliest reports to document the widespread detections of MTBE in New Jersey groundwater. As part of the National Water-Quality Assessment (NAWQA) program, USGS scientists sampled a network of 72 shallow monitoring wells in the Kirkwood-Cohansey aquifer system in the Glassboro area of southern New Jersey. The study area was chosen partly because of its heavy reliance on groundwater, and its rapidly increasing population. The “wells were randomly distributed among agricultural, urban, and undeveloped areas to provide data representative of chemical conditions of groundwater underlying each of these land-use settings.” The wells were sampled in 1996 for a range of nutrients (e.g. nitrate), pesticides and VOCs. Results indicated that MTBE was the second (after chloroform) most frequently detected VOC (at a detection level of 0.1 µg/L) with a detection frequency of 44.4%. Though most VOC detections were low concentrations, MTBE also had the highest maximum concentration detected at 43.8 µg/L. It is also important to note that benzene (the most mobile and most toxic of the gasoline BTEX group) was not among the top 17 VOCs detected in the sampling event, despite a long history of use as part of gasoline.
- **“A Review of Literature for Methyl tert-Butyl Ether (MTBE) in Sources of Drinking Water in the United States”** (*Delzer and Ivahnenko, 2003b*). In a summary of the available literature on MTBE detections in water supplies across the country, USGS scientists summarized findings reported from 13 different states including New Jersey. In one study, domestic supply wells were sampled (on a random basis) from four different regions within the Highlands, Piedmont, and Coastal Plain Provinces of New Jersey. With a reporting limit of 0.1µg/L, MTBE was detected in all four regions with an overall detection frequency of 35.6 percent (37 out of 104 wells sampled). The concentrations

were relatively low ranging from 0.1 to a maximum of 30.2 µg/L (*Delzer and Ivahnenko, 2003*).

- “**Quality of Water from Crystalline Rock Aquifers in New England, New Jersey, and New York, 1995-2007**” (*Flanagan et al., 2012*). This study assessed data collected as part of NAWQA. The data included sampling of 26 domestic wells in a small area with crystalline bedrock in northern New Jersey. The area is part of the “Long Island-New York-New Jersey Coastal Drainages (LINJ) well network.” The wells in New Jersey were sampled in 1997. Out of the 26 wells sampled in the area, 6 wells (23%) had MTBE detections. Five wells (19%) had MTBE detections at concentrations between 1 and 13 µg/L, and one well had a detection above 13 µg/L. Flanagan et al (2012) noted:

“MtBE was detected more often in water samples with apparent ages of less than 25 years than in water samples with apparent ages greater than 25 years. This finding is consistent with the time period of high MtBE use in areas in the United States where reformulated gasoline was mandated.”

#### **Other regional Studies**

As expressed above, the United States Geological Survey (USGS) has conducted several studies on the occurrence of MTBE in water sources across the nation. In these studies the data are collected from a wide array of sources and cover public and private supply wells, rural, urban, deep, and shallow, and from throughout the United States. In many places, these studies have found MTBE to be either the most commonly detected, or the second most commonly detected volatile organic compound (VOC). The widespread detection of MTBE in water sources in the United States is clearly expressed in several USGS reports

- “**Occurrence and Distribution of Methyl tert-Butyl Ether and Other Volatile Organic Compounds in Drinking Water in the Northeast and Mid-Atlantic Regions of the United States, 1993-98**” (*Grady and Casey, 2001*). In this report, USGS scientists examined the presence of VOCs in data supplied by “2,110 randomly selected community water systems (CWSs) in 12 Northeast and Mid-Atlantic States,” including New Jersey, between 1993 and 1998. The study found MTBE to be the most frequently detected VOC in drinking water after the trihalomethane compounds (THMs), compounds that are “potentially formed during the process of disinfecting drinking water with chlorine”.

The study noted that the State with the highest MTBE detection frequency in drinking water was New Jersey, with a detection frequency of 21.5 percent. While VOC data spanned 6 years, the MTBE data collected for New Jersey was limited to two years (1997-1998). Grady and Casey (2001) noted: “With detections reported by 21.5 percent of randomly selected CWSs with data for MTBE, New Jersey reported the greatest frequency of MTBE in drinking water at concentrations at or above 1.0 µg/L, and Virginia (1.3

percent) reported MTBE least frequently.”

This USGS study also found that: “The frequency of MTBE detections in drinking water is significantly related to high-MTBE-use patterns. Detections are five times more likely in areas where MTBE is or has been used in gasoline at greater than 5 percent by volume as part of the oxygenated or reformulated (OXY/RFG) fuels program.” In this study, it was estimated that: “Thirty-four percent of the population served by randomly selected CWSs (community water systems) with data for MTBE may have been exposed to the gasoline additive in their drinking water.”

- **USGS Fact Sheet entitled: “MTBE and other volatile organic compounds- New findings and implications on the quality of source waters used for drinking-water supplies”** (*Zogorski et al., 2001*). In this report, it was concluded that: “MTBE was detected in about 5 percent of ground-water samples collected by NAWQA [National Water Quality Assessment] across the Nation.” …“Specifically, MTBE was detected in about 4 percent of community systems serving less than 10,000 people, and in nearly 15 percent of systems serving greater than 50,000 people.”
- **“Occurrence and Status of Volatile Organic Compounds in Ground Water from Rural, Untreated, Self-Supplied Domestic Wells in the United States, 1986-99”** (*Moran et al., 2002*). This study examined data collected from 1,926 rural domestic wells from 39 states between 1986-99. Of 55 possible VOCs, MTBE again was found to be the second most frequently detected (following trichloromethane), with a detection frequency of 2.2% (compared to 0.3% for benzene).
- **“A National Survey of Methyl tert-Butyl Ether and Other Volatile Organic Compounds in Drinking-Water Sources: Results of the Random Survey”** (*Grady, 2003*). In the random survey, water samples collected from 954 CWSs from all 50 states, Native American Lands, and Puerto Rico, were tested for the presence of 66 VOCs, including MTBE and 3 other gasoline oxygenates. The study was conducted between May 1999 and October 2000. The results showed MTBE detections in 8.7% of the samples (compared to 0.3% for benzene), with MTBE being the second most detected VOC after chloroform, (a THM).
- **“Occurrence and Temporal Variability of Methyl tert-Butyl Ether (MTBE) and Other Volatile Organic Compounds in Select Sources of Drinking Water: Results of the Focused Survey”** (*Delzer and Ivahnenko, 2003*). In the focused survey, water samples collected from 134 CWSs that were suspected or known to contain MTBE, were analyzed for the presence of the 66 VOCs, as above. The study was conducted between September 1999 and April 2001. The results showed MTBE to be the most frequently detected VOC, with a detection frequency of 55.5% (compared to 7.6% for benzene).
- **“Water Resources Data New York Water Year 2002”** (*Spinello et al, 2003*). A USGS report on the water resources data for Long Island included sampling of 50 wells in the four aquifers (Glacial, Jamaico, Magothy, and Lloyd) in Kings and Queens counties, with analysis “for 275 organic and inorganic constituents to assess the ground water as a potable supply” (*Spinello et al., 2003*). Results showed MTBE to be the “most frequently detected contaminant” at 76% detection frequency.

- “**Volatile organic compounds in the Nation’s Groundwater and drinking- Water Supply wells:** Circular 1292” (Zogorski *et al.*, 2006). In this latest USGS study, MTBE was again found to be the second most frequently detected VOC (the first being chloroform) in samples collected from domestic (private) and public drinking water supply wells.
- “**Occurrence of Volatile Organic Compounds in Aquifers of the United States”** (Carter *et al.*, 2008). A study by USGS summarizing the results of analysis for volatile organic compounds in ambient groundwater in aquifers across the United States. The study covered analysis conducted between 1985 and 2002, from 3,498 wells in 98 different aquifer studies. The source of the data included “(1) NAWQA sampling of ground water in aquifer studies during 1993-2002, and (2) data from prior USGS studies and from other Federal, State, and local agencies that sampled ground water in aquifer studies during 1985-1997” (Carter *et al.*, 2008). Each study was based on random sampling, with no specific land-use type being targeted for sampling. Samples were analyzed for 55 volatile organic compounds at assessment levels of 0.02 and 0.2 µg/L. The results of the analysis showed MTBE was the third most frequently detected VOC (at assessment level of 0.2 µg/L) after chloroform and PCE. The study also (Carter *et al.*, 2008) concluded that New Jersey had one of the highest detection frequencies of VOCs in wells (57-60%) emphasizing the vulnerability of the aquifer system to contamination. Carter et al (2008) noted:

“The fate of VOCs in ground water is largely dependent on their persistence under the conditions present in the aquifer. VOCs that are persistent in water are more likely to be detected in ground water because they can travel greater distances from their source before degradation and dilution occur.”

Collectively these studies emphasize the vulnerability of drinking water resources to contamination by MTBE, particularly in light of its relatively short history of production and use. Consider for example, that chloroform was first used as an anesthetic in 1847, and first used for chlorination of a water supply system in the USA in 1908, while MTBE was first approved for use as an octane booster by the USEPA in 1979 (Zogorski *et al.*, 2006). Given the relatively short history of use of MTBE in the United States, such widespread detections of MTBE in water sources nationwide is alarming.

In testimony before the U. S. House of Representatives (November 1, 2001), Robert Hirsch, the Associate Director for Water for the USGS stated that:

“Through our interpretations of this large data set we have also determined that low-levels of

MTBE are detected in about 1 out of 5 wells in MTBE high-use areas. Although we do not expect to see great change in these results over time, we recognize that there may be a delay in the detection of MTBE in some wells-particularly those that are deeper and may be farther from the source of contamination. MTBE is the second most frequently detected volatile organic compound (VOC). Chloroform, a drinking-water disinfection by-product and a commercial solvent, is the most frequently detected VOC.”

In other words, MTBE, a compound whose widespread use only began relatively recently, was already detected in 20% of wells in high-use areas and was the second most frequently detected VOC in 2001. Further, this contamination was expected to migrate deeper over time, eventually impacting deep water supply wells. These facts suggest that we now are only beginning to see the initial developments of potential groundwater quality impacts due to widespread MTBE contamination. I must reiterate that our past experience with other groundwater contaminants demonstrates that the full impacts from compounds like MTBE evolve over a decadal time scale, not over a mere few months or years, in groundwater basins like those upon which California and other states depend for drinking water supply.

## **9 Ethanol**

Alcohols were first added to gasoline as octane boosters in the 1930s, and ethanol blended gasoline was sold in the Midwest in the 1930s and 1940s (*Moyer, 2003*). The use of ethanol as an alternative, or renewable, energy source was stimulated in the mid 1970s, in response to the oil embargoes of 1973 and 1979 (*CRS, 2003*). More recently, with the introduction of the Clean Air Act Amendments of 1990, ethanol was also used as a fuel oxygenate, though mostly in the Midwestern states (Table 7.1). In 1998 it was the “second most common” fuel oxygenate present in about 15% of oxygenated gasoline (*USEPA 2004*). With the ban or phase-out of MTBE in many States, the production of ethanol, a replacement oxygenate, has risen sharply, doubling in a matter of few years (*Zhang et al., 2006*). Yet, despite a long history of use, surprisingly little field data is available on environmental impacts of ethanol-blended gasoline.

As a gasoline oxygenate, less ethanol by volume (compared to MTBE) is needed to achieve the oxygen requirements necessary to meet the Amendments to the Clean Air Act. For Oxygenated Fuel (2.7% by weight oxygen) 7.3% ethanol by volume would be added to gasoline, compared to concentrations of up to 15% by volume MTBE (*USEPA 2008*). For Reformulated Gasoline (2% by weight oxygen) 5.4% ethanol by volume would be added to gasoline, compared to 11% by volume MTBE (*USEPA 2008*).

Some recent research has suggested that at some sites, the use of ethanol might increase groundwater contamination relative to the use of conventional gasoline. However, and as will be discussed next, studies have concluded that the impacts from ethanol to groundwater are expected to be “less problematic” (*NEIWPCC and NESCAUM, 2001*) and “more manageable” (*Powers et al., 2001b*) compared to MTBE, and that ethanol is “an acceptable substitute” to MTBE (*Dakheel et al., 2003*).

## 9.1 Properties of Ethanol in Soil and Groundwater

Ethanol is miscible in water, and therefore tends to partition preferentially into water from gasoline. From its pure phase in contact with water, ethanol can reach aqueous concentrations that are many times those of the maximum aqueous concentration attainable by MTBE. Once in water, ethanol is considerably less volatile than MTBE.

Laboratory and field studies indicate that ethanol is easily, and preferentially (relative to BTEX), biodegraded by indigenous microorganisms in groundwater under different conditions (*Corseuil and Alvarez, 1996; Alvarez et al., 2001; Powers et al. 2001b; Deeb et al., 2002; Ruiz-Aguilar et al., 2002; Mackay et al., 2006; Zhang et al. 2006*).

Because of its ability to biodegrade easily, ethanol itself “is unlikely to be a significant groundwater pollutant” (*Powers et al., 2001b*). For example, only 5 states require sampling and analysis for ethanol in groundwater at gasoline release sites, compared to 31 states that require sampling and analysis for MTBE (*NEIWPCC, 2003*). States that use mostly ethanol as oxygenate, such as Minnesota and North Dakota, consider the risks to be low because ethanol degrades easily as indicated by a survey of the 50 states experience with oxygenates at LUST sites, conducted by the New England Interstate Water Pollution Control Commission (*NEIWPCC, 2003*). For example the responses from both Minnesota and North Dakota indicated that the oxygenate used in those states (ethanol) is “not perceived to be a problem”. The Minnesota response indicated “the human risks associated with ethanol are very low. Also, ethanol degrades very quickly.” North Dakota’s response was similar: “ethanol will degrade naturally if released into the environment.”

## 9.2 Cosolvency and Mobility Effects of Ethanol on BTEX

At concentrations exceeding those required for RFG, ethanol in gasoline can have concentration-dependent cosolvency and mobility effects. Co-solvency effects are thought to arise from a reduction in the polarity of water, effecting the partitioning of BTEX compounds between NAPL, water, soil, and vapor phases and resulting in higher equilibrium, aqueous concentrations of BTEX (*Heermann and Powers, 1998; Da Silva and Alvarez, 2002*).

Research indicates that significant cosolvency effects are not likely to arise from a spill of ethanol oxygenated gasoline (up to 10% by volume ethanol (*Da Silva and Alvarez, 2002; Heermann and Powers 1998; Powers and Heermann, 1999*). However, higher concentrations, e.g., 50% by volume ethanol in gasoline, can lead to the free migration of benzene, toluene, and o-xylene (BToX) within the local groundwater (*Da Silva and Alvarez, 2002*). This raises concerns regarding neat ethanol spills, e.g., from bulk storage facilities, in the presence of other petroleum hydrocarbons (*Powers and McDowell, 2001*), like the one that occurred at the Pacific Northwest Terminal site spill described later.

Ethanol in gasoline can also affect the mobility of BTEX compounds through a reduction in the retardation of BTEX and the surface tension of water (*Heermann and Powers, 1998; Da Silva and Alvarez, 2002; Powers and McDowell, 2001*). These effects are seen as having potentially positive and negative impacts on groundwater quality. On the one hand, increases in NAPL mobility in the unsaturated zone may affect leaching of contaminants to groundwater. On the other hand, increased mobility would likely improve the efficiency of some remediation efforts (*Powers and McDowell, 2001*). Even in the presence of cosolvency and mobility effects, both ethanol and BTEX compounds are expected to attenuate through biodegradation as explained in the following section.

### 9.3 Inhibitory effects of Ethanol on the Biodegradation of BTEX Compounds

Microbial degradation appears to be the dominant natural attenuation mechanisms for ethanol and BTEX in groundwater (*Alverez et al., 2001*). To carry out the biodegradation of ethanol and BTEX, indigenous microorganisms rely on other constituents referred to as electron acceptors. Because ethanol is more easily biodegraded than BTEX, the preferential biodegradation of ethanol can interfere with the biodegradation of BTEX compounds by depleting electron acceptors used for BTEX biodegradation (*Corseuil et al., 1996*). Still, effects of the presence of ethanol on BTEX degradation can vary. For example, laboratory experiments have found that ethanol can actually enhance toluene degradation in the presence of excess electron acceptors (*Alverez et al., 2001*). Similarly, ethanol, at relatively low concentrations, can enhance benzene degradation under denitrifying conditions (*Wu et al., 2009*).

As discussed in the following sections, field and modeling studies suggest that for a large enough spill that results in significant ethanol and BTEX groundwater plumes, the effects caused by the preferential biodegradation of ethanol in groundwater can either increase the length of, or prolong the duration of, BTEX plumes (*Alverez et al., 2001; Buscheck et al., 2001; Mackay et al., 2006*). On the other hand, natural attenuation (biodegradation) in the unsaturated zone, or locally in groundwater, may virtually prevent the spread of ethanol and BTEX contamination from small liquid or vapor phase releases of ethanol-oxygenated gasoline (10% by volume) (*Dakhel et al., 2003*). Furthermore, because ethanol and BTEX are relatively easy to biodegrade, engineered remediation (introduction of oxygen, nutrients) can be very effective at attenuating contamination and preventing its spread to sensitive receptors (*Heaston et al., 2010*).

The following field and modeling studies illustrate some of the effects of ethanol addition to gasoline that are mentioned above.

### 9.4 Field Studies

#### **Small Volume Releases** (*Dakhel et al., 2003*)

To mimic a small-volume gasoline spill, a controlled experiment involving the release of an artificial gasoline containing MTBE and ethanol (at 5% by weight of each) was performed in a lysimeter having a 2.3 m thick vadose zone of alluvial sand overlying a gravel aquifer. The experiment consisted of three sequential phases. In the first phase of 41 days, no recharge water was applied to the surface. During this phase, MTBE and volatile gasoline hydrocarbon vapors migrated by diffusion to, and accumulated in, groundwater. Ethanol, on the other hand, naturally attenuated in the vadose zone and did not accumulate in groundwater. During the second phase of 30 days, 5mm/day recharge was applied to the surface over a period of 30 days to simulate infiltration due to a rain event. During this phase, MTBE, ethanol, and soluble gasoline constituents (e.g., BTEX) were flushed to the groundwater. During the third phase of 120 days, no recharge was applied. Once in groundwater, ethanol, benzene and other detectable petroleum hydrocarbons (with the exception of isoctane) were rapidly biodegraded. More than 99% of the ethanol disappeared. In contrast, there was no evidence of MTBE biodegradation. MTBE reached peak concentrations of more than 250,000 µg/L in groundwater and persisted at concentrations greater than 125,000 µg/L

for the duration of the experiment lasting about 6 months. Higher MTBE concentrations would be expected to result from a spill of oxygenated gasoline with 11% by volume MTBE. The study's authors concluded that: "groundwater quality will be affected considerably less at small volume release sites when MTBE-free gasoline or gasohol [ethanol blended gasoline] are spilled" and that "MTBE-free gasoline would be less harmful for groundwater resources and that ethanol is an acceptable substitute."

These results have important implications for the small volume vapor- and liquid-phase releases that beset USTs and UST systems, including upgraded 1998 compliant ones. Ethanol in small volume releases at a gasoline station is likely to bioattenuate significantly in the vadose zone and, in the event that it reaches the water table, locally within groundwater. In contrast, MTBE is expected to migrate to, and persist in, groundwater.

### **Neat Ethanol Spill Site at the Pacific Northwest Terminal Refinery** (*Buscheck and O'reilly, 2001*<sup>32</sup>; *Powers and McDowell, 2001*)

In March 1999, a 19,000-gallon (about 72,000 liters) release of neat ethanol from an above ground storage tank at the Pacific Northwest Terminal Refinery produced an ethanol plume that ultimately extended more than 250 ft (76 m) down-gradient. While the ethanol plume appeared to degrade, the spill affected NAPL behavior and aqueous phase benzene concentrations of the preexisting hydrocarbon contamination on site. Benzene concentrations increased by as much as a factor of 15 and the LNAPL thickness in many of the wells increased. Both observations may have been due to cosolvency effects and the inhibition of biodegradation due to the spill of (pure) neat ethanol.

### **Transport of Ethanol in a Sand and Gravel Aquifer** (*Zhang et al., 2006*).

This study monitored plumes originating from an injected pulse containing approximately 220 mg/L ethanol and 16 mg/L bromide in a sand and gravel aquifer for 2.5 months. Observations suggest that ethanol migration was attenuated only through biodegradation. Ethanol rapidly degraded with a computed half-life of 2.2 days.

### **Site 60, Vandenberg Air Force Base, CA** (*Mackay et al., 2006*)

Two experiments were performed side-by-side, simultaneously, over a period of 9 months in an aquifer (with sulfate reducing conditions) underlying a former fuel station to evaluate the effects of ethanol on the biodegradation of BToX (benzene, toluene and oxylene). On one side, BToX was continuously injected; on the other side, BToX with ethanol was continuously injected. The BToX plumes on both sides migrated approximately the same distance and then retracted. The BToX plumes on the side with no ethanol retracted at a significantly greater rate, and further, than the BToX plumes on the side with ethanol. The researchers hypothesized (1) that the plumes reached the same length initially due to an acclimation period before biodegradation began, and (2) that the slower retraction of BToX on the ethanol side was due to the preferential biodegradation of ethanol and associated depletion of dissolved sulfate, leading to methanogenic/ acetogenic conditions less effective for biodegradation of BToX. These results provide field evidence for predicted increases in BTEX plume lengths as a result of ethanol addition to gasoline.

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<sup>32</sup> <http://www-erd.llnl.gov/ethanol/proceed/terminl.pdf> Buscheck and O'reilly, 2001 last accessed 2/27/07

## **LUFT sites in Iowa and Kansas (*Ruiz-Aguilar et al., 2003*).**

This study compared the lengths of plumes originating from ethanol-oxygenated gasoline (10% ethanol by volume) and conventional gasoline LUFT sites in Kansas and Iowa, respectively. The authors concluded that plumes for benzene in ethanol-containing gasoline were significantly longer than plumes for benzene in conventional gasoline. The same relationship was not found for toluene plumes. Median benzene plume lengths were found to be 36% longer for ethanol-containing versus conventional gasoline sites. These data are consistent with the expected inhibitory effects of biodegradation indicated by the modeling studies discussed below. Even so, caution is warranted when comparing plumes lengths between different geologic settings and from sites that are not well monitored.

## **9.5 Modeling Studies**

Researchers have developed mathematical models aimed at studying the effects of ethanol in gasoline (10% by volume) on plume behavior and the size of BTEX groundwater plumes (*Dooher, 1999; McNab et al., 1999; Molson and Barker, 2000; Powers et al., 2001a; McNab, 2001; Molson et al., 2002; Deeb et al., 2002*). Overall, these models predict increases in BTEX plume lengths due to the addition of ethanol, a result consistent with some of the field studies discussed above. Maximum benzene plume lengths originating from conventional gasoline spills at LUFT sites are typically several hundred feet in length (*Fogg et al., 1998b; Happel et al., 1998*). Results from these models predict that ethanol may increase benzene plume lengths from less than 10% to as much as 150% depending on the modeling approach and specified subsurface conditions. The approaches used in many of these studies are generally considered to be conservative in that they rely on a number of simplifying assumptions that, if incorrect, would tend to overestimate the effects of ethanol on BTEX plume lengths (*Powers et al., 2001a*).

In addition, in all of these modeling studies it is assumed that ethanol and BTEX compounds freely migrate as a homogenous mixture from the unsaturated-zone to the water table. However experiments suggest that in the unsaturated zone, ethanol migration is preferentially attenuated relative to BTEX (*Powers and McDowell, 2001*). This phenomenon could increase the time available for the biodegradation of ethanol in the unsaturated zone and decrease the rate at which ethanol leaches to the saturated groundwater. In this case, less ethanol would reach groundwater leading to potentially smaller effects on the lengthening of BTEX plumes than predicted by the modeling studies discussed above. Accordingly, *Powers and McDowell (2001)* state:

“Attempts to mathematically model the overall fate of ethanol and BTEX in the subsurface must incorporate the mechanisms seen in these experiments in order to more accurately depict the mechanisms that affect the initial distribution of ethanol and gasoline in the unsaturated zone. The results presented here suggest that the retention of ethanol in the unsaturated zone may be significant.”

## **9.6 Summary**

As discussed previously, the most important factor contributing to the differential fate of MTBE and ethanol in the

subsurface appears to be biodegradability. The poor biodegradability of MTBE allows for long persistent plumes, exposing larger volumes of water to contamination. A detailed review of the health and environmental impacts of the addition of ethanol to gasoline in the Northeast (*NEIWPCC and NESCAUM, 2001*), jointly prepared by the New England Interstate Water Pollution Control Commission (NEIWPCC) and the Northeast States for Coordinated Air Use Management (*NESCAUM, 2001*) concluded with recommendations for removal of the oxygen mandate for reformulated gasoline. However, in the absence of a removal, the report concluded that: “from a water quality protection standpoint, the potential adverse impacts associated with ethanol are clearly less problematic than for MtBE-blended gasoline.” (*NEIWPCC and NESCAUM, 2001*). Scientists at Lawrence Livermore National Laboratory (*Powers et al., 2001b*) reached similar conclusions: “given the choice between MTBE and ethanol, and with our current level of understanding, it appears that the groundwater resource impacts associated with the use of ethanol will be less significant and more manageable than those associated with MTBE”.

Evidence from the state of Minnesota, where the use of ethanol in gasoline at 10% (E10) was mandated in 1997 (*Toso, 2009*), bear out the above predictions. In a recent article on the use of ethanol in gasoline in Minnesota, *Toso* (2009) from the Minnesota Pollution Control Agency (MPCA) noted that: “after an informal review, the potential differences between an E10 and an ethanol-free gasoline release were deemed minor and covered under existing investigation and cleanup policy, which is based on groundwater plume delineation. No ethanol specific issues have arisen from E10 releases since then.” (*Toso, 2009*). Furthermore, it should be noted that although ethanol has been in use in gasoline since the 1930s, to my knowledge, there is no evidence to date that it has posed a problem as a contaminant in groundwater drinking supplies.

Based on all of the above, and given the dispersed nature of fuel contamination, it is clear that ethanol (at 5.4% requirement in reformulated gasoline) would have been a much better choice as an oxygenate in gasoline, from a groundwater protection standpoint.

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## 11 Glossary of Terms

**Acclimation period:** The time elapsed between the introduction of contaminant(s) and observation of microbial activity (biodegradation).

**Activated Carbon:** A highly adsorbent form of carbon used to remove odors and toxic substances from liquid or gaseous emissions.

**Activated Sludge:** sludge particles (flocs) produced by the growth of large groups of organisms (mostly bacteria, but also fungi and protozoa) during the secondary treatment process of wastewater. This process involves agitation of the liquid and aeration, to maintain high oxygen concentration to maximize microbial activity.

**Alluvial aquifer:** A geologic formation of alluvium (materials such as clay, silt, sand, and gravel deposited by streams or rivers) that can yield significant quantities of water to wells or springs.

**Bioaugmentation:** the addition of bacteria or other microorganisms that can biodegrade a specific organic compound.

**Biobarrier:** a biologically active zone installed in the subsurface, in a direction perpendicular to that of the normal flow of contaminated water, to allow for the biodegradation of the contaminant.

**Biodegradation:** biologically induced structural transformation of a chemical compound.

**Bioremediation:** the use of microorganisms in removing contaminants from water, soil or air.

**BTEX:** benzene, toluene, ethyl benzene and xylene (*m*-, *o*-, and *p*- xylene). These volatile organic compounds are of particular concern in gasoline, because of their water solubility and toxicity.

**BToX:** benzene, toluene, and o-xylene.

**CDHS:** California Department of Health Services

**CDWR:** California Department of Water Resources

**Cosolvency:** An effect wherein the presence of more than one organic contaminant causes a change in the partitioning behavior of one or more of these contaminants, as compared to the condition where they are present alone.

**CWS:** community water supply (Johnson et al., 2000); community water system (*Water-Resources Investigations Report WRIR00-4228*, 2001)

**Diffusion:** the movement of a chemical resulting the random motion of molecules.

**Dilution:** reduction of contaminant concentration by mixing with less-contaminated, or uncontaminated, water.

**DLR:** detection limit for reporting purposes (DLR) of 3 µg/L. The DLR is the level at which DHS is confident about the quantity being reported. Results at or above the DLR are required to be reported to DHS; some laboratories may report results at lower concentrations. As defined in the CDHS web page.

(<http://www.dhs.cahw.net/ps/ddwem/chemicals/MTBE/mtbeindex.htm>)

**Enzyme:** a protein produced by a living organism, which functions as a biochemical catalyst, facilitating a specific chemical reaction.

**Fractured medium:** A rock or clay formation which is mostly solid but contains cracks that can transmit or store water.

**GAC:** granular activated carbon. A highly adsorbent form of carbon (possessing a high porosity and high surface activity) typically used to remove organic contaminants from water.

**Half-life:** the time required for the initial concentration of a compound to be reduced by half, often used in reference to radioactive decay, and sometimes applied to biodegradation and other reactions.

**Humic substances:** material derived from the degradation of plants algae and other organisms, which accounts for much of the naturally occurring organic carbon<sup>33</sup>.

**Inoculum:** material that serves as a source of microbial cells, such as activated sludge, soil, or sediment

**Intrinsic biodegradation:** biodegradation of contaminants by indigenous microorganisms.

**LUFT:** leaking underground fuel tank

**LUST:** leaking underground storage tank

**Lysimeter:** a device used for measuring the percolation of water through soil, and for determining the soluble constituents removed through drainage.

**MCL:** maximum contaminant level, refers to “enforceable regulatory standards under the Safe Drinking Water Act and must be met by all public drinking water systems to which they apply” (from:

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<sup>33</sup> Scott et al., 1998

<http://www.dhs.ca.gov/ps/ddwem/chemicals/MCL/mclindex.htm>). In New York, the MCL for MTBE is 10 µg/liter. In some States, a primary MCL is set on health-based standards, while a different secondary MCL is set on aesthetic standards such as taste and odor. In California, the secondary MCL for MTBE is 5 µg/liter, while the primary MCL for MTBE is 13 µg/liter.

**Microcosm:** a laboratory-scale model used to study the interactive relationships of microbial communities and the ability to biodegrade contaminants, among other things.

**Mineralization:** the complete biodegradation of an organic compound into carbon dioxide, water, inorganic ions and molecules, and possibly cellular material.

**The MTBE Blue Ribbon Panel:** “was created by a Charter from the Clean Air Act Advisory Committee (CAAAC) to provide independent advice and counsel to EPA on policy issues associated with the use of MTBE and other oxygenates in gasoline” (<http://www.epa.gov/oms/consumer/fuels/oxypanel/blueribb.htm>).

**Monooxygenase:** an enzyme that catalyzes the addition of one atom of molecular oxygen to a molecule to be transformed.

**PADD:** petroleum administration defense district.

**ppb:** parts per billion US; parts per 1,000 million; microgram per liter, µg/l.

**PWS:** public water supply.

**Recalcitrant:** resistant to microbial degradation.

**RFG:** reformulated gasoline

**SWRCB:** State Water Resources Control Board

**Trihalomethanes (THM):** Disinfectant by-products formed when chlorine reacts with organic compounds in water. These include trichloromethane (chloroform) dibromochloromethane, bromodichloromethane, and tribromomethane (bromoform).

**USEPA:** United States Environmental Protection Agency.

**USGS:** United States Geologic Survey, defined on the web site (<http://www.usgs.gov/>) as: “Federal source for science about the Earth, its natural and living resources, natural hazards, and the environment.”

**UST:** underground storage tank.

**VOC:** volatile organic chemical.

## **Appendix A: C.V. of Dr. Graham E. Fogg**

## Curriculum Vitae

### **Graham E. Fogg**

*Hydrology Program, Department of Land, Air, and Water Resources  
Hydrologic Sciences Graduate Group  
One Shields Avenue  
University of California, Davis 95616  
(530) 752-6810 FAX: (530) 752-5262  
E-mail: gefogg@ucdavis.edu*

#### **ACADEMIC BACKGROUND**

B.S. Hydrology, University of New Hampshire, 1975  
M.S. Hydrology, University of Arizona, 1978  
Ph.D. Geology, The University of Texas at Austin, 1986

#### **HONORS**

Phi Kappa Phi Honor Society  
Geological Society of America Birdsall-Dreiss Distinguished Lecturer, 2002  
Fellow, Geological Society of America  
O.E. Meinzer Award, 2011

#### **AREAS OF EXPERTISE**

- A. Contaminant transport in complex porous media; Geologic/geostatistical characterization of subsurface heterogeneity
- B. Numerical modeling of ground-water systems
- C. Regional system hydrogeology: ground-water hydraulics, sustainability of groundwater quality, groundwater and surface water interaction

#### **PROFESSIONAL EXPERIENCE**

- A. Present Position: Professor of Hydrogeology, Hydrology Program, Department of Land, Air, and Water Resources, University of California, Davis (July 1, 1996 - present). Vice Chair for Hydrology 1998-2001.
- B. Associate Professor of Hydrogeology, Hydrology Program, Department of Land, Air, and Water Resources, University of California, Davis (March 15, 1989 - June 30, 1996).
- C. Research Associate, Bureau of Economic Geology, The University of Texas at Austin. (Sept. 1985 - Jan. 1989).

Research multiphase flow hydrodynamics of hydrocarbon reservoirs in West Texas and stochastic methods of characterizing reservoir interconnectedness. Research regional hydrogeologic processes as related to potential impacts of lignite mining on water quality and quantity. Develop improved methods of describing aquifer heterogeneity using geologic information.

- D. Research Scientist Associate, Bureau of Economic Geology, The University of Texas at Austin (January 1978 - August 1985).

Conduct research on hydrogeologic suitability of salt domes in the East Texas Basin for storage

of high-level nuclear wastes. Assemble and interpret hydrogeologic data bases; design and supervise hydrologic test-drilling programs; construct detailed two and three dimensional ground-water flow models for analyzing near-dome flow systems and potential radionuclide pathways to the biosphere; research new methods of modeling mass transport and of describing aquifer heterogeneity.

## PROFESSIONAL SOCIETIES

American Geophysical Union, Water Resources Division  
Association of Ground Water Scientists and Engineers  
International Association of Hydrogeologists  
Geological Society of America  
Groundwater Resources Association of California

## SELECTED PROFESSIONAL ACTIVITIES (last 10 years)

Manuscript reviewer, Hydrogeology Journal, Ground Water journal, Water Resources Research, Mathematical Geology, Advances in Water Resources, Transport in Porous Media, among others.  
Proposal reviewer, National Science Foundation  
Member, Peer Review College of the Danish Council for Strategic Research  
Member, Committee to evaluate research performance of GEUS (Geological Survey of Denmark and Greenland), Sept.-Nov., 2007  
Co-Chair of Subsurface Characterization Panel, Dept. of Energy, Basic Research Needs Program in Subsurface Science (to determine basic research needs for the future, concerning subsurface CO<sub>2</sub> sequestration and nuclear waste isolation), 2007-08.

## SELECTED UNIVERSITY ACTIVITIES

College of Agricultural and Environmental Sciences Academic Planning Committee, 2009.  
Member, Search Committee for Director of John Muir Institute for the Environment  
Chair, Search Committee for Associate/Full Cooperative Extension Specialist and Associate Full Professor in Water Resources (Robert Hagan Endowed Chair), 2008 – 2009.  
Member, delegation to University of Concepcion, Chile, to develop collaborative water programs, March – present 2009.  
Vice Chair for Hydrology, Department of Land, Air and Water Resources, 1998 – 2001.  
Chair, Hydrologic Sciences Graduate Group, 1993 – 1998, 2006 – present.  
Graduate Advisor, Hydrologic Sciences Graduate Group, 1991 – 1993, 1998 – 2000, 2001–present.  
Graduate Advisor, Water Science Graduate Group, 1989 – 1991.

## COURSES TAUGHT

The University of Texas at Austin Geology Dept.: Groundwater Modeling, 1987.

UC Davis: WSC 298, Contaminant Transport Processes, 1989 (graduate).

WSC 149B, HYD 145B, Groundwater Hydrology, 1990, 1991, 1992, 1993, 1994, 1996, 1997 (undergraduate).

HYD 10, Water and Power, 2001, 2003, 2004, 2006, 2007, 2008, 2009.

HYD 146, Hydrogeology and Contaminant Transport, 1998, 1999, 2000, 2001, 2003, 2004,

2007, 2008, 2009.

HYD 269, Numerical Modeling of Groundwater Systems, 1990, 1991, 1992, 1993, 1994, 1996, 1997, 1998, 1999, 2000, 2001, 2004, 2006, 2008 (graduate).

HYD 273, Introduction to Geostatistics, 1990, 1992, 1994, 1996, 1998, 2000, 2002, 2004, 2006, 2008 (graduate).

## **SELECTED SHORT COURSES AND WORKSHOPS DEVELOPED AND TAUGHT**

"Modeling of Heterogeneity and Transport in Geologic Media: Transition Probability, Markov Chains and Random-Walk Particle Methods (TProGS & RWHET)" June 22-27, 2009. University of Copenhagen, Denmark.

"Hydrofacies Modeling with TProGS, a Geostatistical Approach based on Transition Probability and Markov Chains." February 6-9, 2006. U.S. Geological Survey, Sacramento State University, Sacramento, CA.

"Hydrofacies Modeling with TProGS, a Geostatistical Approach based on Transition Probability and Markov Chains." October 20-24, 2003. Geocenter Copenhagen, Denmark.

"Principles of Groundwater Flow and Transport Modeling." 3-day short course.

Sponsor/Coordinator: University Extension, University of California, Davis. Taught one to two times per year since 1998.

"The Groundwater Workshop." 1-day workshop. Sponsor/Coordinator: The Water Awareness Committee of Monterey County, Inc. March 19, 1998. Salinas, CA.

"Applied Groundwater Hydrology: Principles, Measurements, and Interpretation." 2-day short course. Sponsor/Coordinator: Cal/EPA Department of Toxic Substances Control, CA Groundwater Resources Association and the University of California, Davis. May 19-20, 1998. Berkeley, CA.

"Introduction to Applied Groundwater Hydrology: Principles, Measurements, and Interpretation." 3-day short course. Sponsor/Coordinator: University of California Davis. November 6-7, 1996; Fresno County, CA; November 1997, Sacramento, CA.

## **SELECTED EXPERT TESTIMONY (public service and litigation)**

- Expert witness and testimony in Imperial Irrigation District vs. Coachella Valley Water District, 1987-1988.
- Testimony to California Assembly Committee dealing with registration of groundwater professionals. 1991.
- Testimony to California Joint Legislative Audit Committee. "Is Groundwater Quality Falling Through the Legislative Cracks?" February 9, 1999.
- Testimony at CalEPA Public Hearing on MTBE in California, Diamond Bar, CA. Impacts of MTBE on California Groundwater. February 19, 1999.
- Testimony at CalEPA Public Hearing on MTBE in California, Sacramento, CA. Impacts of MTBE on California Groundwater. February 23, 1999.

- Testimony to EPA Blue Ribbon Panel on MTBE Issues. Impacts of MTBE on California Groundwater. March 25, 1999.
- Expert witness and testimony in South Lake Tahoe Public Utility District vs. ARCO et al., 2000-2001.
- Expert witness and testimony in Citizens for a Better Environment vs. Union Oil Corp. et al., 2000-2001.
- Expert witness and testimony to International Tribunal in the Arbitration Under Chapter 11 of the North American Free Trade Agreement (NAFTA) Between Methanex Corporation (Canada) and The United States of America, World Bank, Washington, D.C., June 11, 2004.
- Expert witness and testimony to arbitration panel in City of Santa Monica vs. Shell Oil Co. et al., 2005.
- Expert witness and testimony in Fruitridge Vista Water Company vs. ARCO et al., 2007.
- Expert witness and testimony in County of Suffolk and Suffolk County Water Authority vs. Amerada Hess Corp., et al., and United Water New York vs. Amerada Hess Corp., et al., 2008.
- Expert witness and testimony in *City of New York v. Amerada Hess Corp., et al.* 04 Civ. 3417, 2009.

#### **INVITED TALKS AND LECTURES (2000-present)**

Dichotomy of Scale Concepts in Geology and Hydrology, Aquifer Sedimentology Conference (SEPM special conference), Sante Fe, Sept. 29, 2000.

Heterogeneity, the Pumping Test and the Plume: An Evolution (or Devolution?) of Hydrogeologic Concepts, Michigan State University, Oct. 19, 2000.

Geologic Heterogeneity and Predictive Uncertainty in Hydrogeologic Models, AGU, Dec. 15, 2000.

#### ***2002 Geological Society of America Birdsall-Dreiss Distinguished Lectures***

**Talk A:** Plume Behavior in Heterogeneous Geologic Systems: Natural Attenuation, Remediation, and the Role of Diffusion

**Talk B:** Groundwater Vulnerability and the Meaning of Groundwater Age Dates

**Talk C:** A Geologic Approach to Simulation of Subsurface Hydrology

Total institutions visited: 45 (37 Universities; 2 USGS offices; 1 USGS workshop; 2 state surveys; 1 GS of Canada office; 1 national lab; 1 consulting firm; 1 state professional society)

Total lectures delivered: 56

All lectures were by invitation.

Date of Lecture	Location	Talk
1. January 14	Indiana University (Bloomington)	B, C
2. January 15	Wright State University (Dayton)	C, A
3. January 16	University of Kentucky, Lexington	B
4. January 17	Ohio State University (Columbus)	C
5. January 18	University of Akron	B
6. January 28	University of Mississippi (Oxford)	C
7. January 29	University of Memphis	B
8. February 1	University of Tennessee, Knoxville	A, B
9. February 5	Clemson University	A
10. February 6	Georgia State University (Atlanta)	A
11. February 12	Fresno State University	B
12. February 19	Boise State University	C
13. February 21	University of British Columbia	B
14. February 25	Louisiana State University (Baton Rouge)	B
15. February 27	Texas A&M University	A
16. March 1	University of Texas at Austin	A
17. March 4	Bureau of Economic Geology	B
18. March 4	Intera, Inc.	C
19. March 12	Penn State University	B
20. March 13	MIT	B
21. March 14	University of New Hampshire	B, A
22. April 2	Iowa State University	A, B
23. April 3	University of Wisconsin	A, B
24. April 9	University of Waterloo	B
25. April 10	Syracuse University	C
26. April 11	Canadian Geological Survey (Ottawa)	C
27. April 23	Northern Arizona University	B
28. April 24	USGS mtg. Tucson	C
29. April 24	University of Arizona Ty Ferre	A
30. April 30-May 1	New Mexico Tech	C, B
31. May 2	Los Alamos National Laboratory	A
32. May 3	University of New Mexico (Albuquerque)	C, B
33. September 13	University of Nevada Reno	B
34. September 16	Johns Hopkins University	B
35. September 17	Delaware Geological Survey, U. of Delaware	C
36. September 19	University of Virginia	A, B
37. September 23	USGS – Reston	B
38. October 4	University of Hong Kong, China	B
39. October 7	University of Chang'an, Xian, China	B
40. October 10	Chinese University of Geosciences, Beijing, China	B
41. October 16	Stanford University	A
42. October 17	USGS – Menlo Park	C
43. October 18	University of California, Santa Cruz	B
44. October 29	GSA Denver.	B
45. November 11-13	University of Hawaii	A, B, C

Influence of Paleosols on Fluid Flow and Transport: Perspective on Alluvial Complexity and Hydrogeology, Presentation before the DOE Nuclear Waste Technical Review Panel, Las Vegas, NV, Jan. 28, 2003.

Review of IGSM, and Perspective on Future Incarnations, Annual Meeting of CA Bay-Delta Modeling Forum, Asilomar, CA, Feb. 2003.

Hydrostratigraphy and Contaminant Transport Beneath Rancho Cordova and Vicinity.... Plus Discussion of Cosumnes Phenomena, American River Conference, CSU Sacramento, Nov. 2003.

Perspective on Heterogeneity, Groundwater Monitoring, and Natural Attenuation, Geological Society of America Annual Meeting, Nov. 2003, Seattle, WA.

Perched Aquifer Hydrology: An Unexplored Frontier?, Geological Society of America Annual Meeting, Nov. 2003, Seattle, WA.

Scientific Perspective on Groundwater, California Water Forum (Jay Lund), April 14, 2004.

Analysis of Human Exposure to Perchlorate Through Coupled Modeling of Groundwater and the Surface Distribution System, US EPA Superfund Conference, Oct. 19, 2004, Sacramento, CA.

Fish Passage, Groundwater and Base Flow - The Case of the Cosumnes River. CALFED Science Conference, Oct. 2004, Sacramento, CA.

The Stochastic Facies Model of Heterogeneity: A Necessary Tool or Unnecessary Tedium?, Geological Society of America Annual Meeting, Nov. 2004, Denver, CO.

The Role of Perched Aquifers in Hydrological Connectivity and Biogeochemical Processes in Vernal Pool Landscapes, Great Central Valley, California. Presentation to Sacramento Parks and Recreation Board, Nov. 2004.

Contaminant Transport in Groundwater, With Special Focus on MTBE, HYD 198 Lecture (Peter Hernes' class), Feb. 24, 2005.

Contaminant Transport in Groundwater, With Special Focus on MTBE, HYD 198 Lecture (Peter Hernes' class), Mar. 3, 2005.

**Keynote:** Washington Hydrogeology Symposium, April 14, 2005, Tacoma, WA: **Groundwater Vulnerability and the Meaning of Age Dates.**

Recent Perspectives on Groundwater Age: Estimation, Modeling, and Water Quality Sustainability, 25<sup>th</sup> Biennial Conf. & CA Groundwater Resources Association Meeting, Oct. 2005, Sacramento, CA.

The Hydrofacies Approach and Why  $\ln K \sigma^2 < 5-10$  is Unlikely, American Geophysical Union Fall Meeting, Dec. 2004.

Groundwater Quality Sustainability, Creeping Normalcy, and a Research Agenda, Geological Society of America Annual Meeting, Oct. 17, 2005, Salt Lake City, UT.

Contaminant Transport in Groundwater, With Special Focus on MTBE, SAS 8, Jan. 2006.

**Keynote:** Computational Methods in Water Resources Conference, June 19, 2006, Copenhagen, Denmark: If Heterogeneity Is So Important, Why Do We Still Ignore It?

Practical Approach For Dealing With Heterogeneity and Why It Is Essential for Modeling Transport, California Groundwater Resources Association Conference on High-Resolution Site Characterization and Monitoring, November 15, 2006.

Contaminant Hydrogeology: Rejuvenate the Research Agenda, Geological Society of America Annual Meeting, Oct. 23, 2006, Philadelphia, PA.

**Keynote:** If Heterogeneity Is So Important, Why Do We Still Ignore It?, DOE Environmental Remediation Science Program, Oct. 23, 2006, Oak Ridge TN.

Contaminant Transport in Groundwater, With Special Focus on MTBE, SAS 8 Guest Lecture, Jan. 2007.

What's in a Plume Model and Why Should We Care?, Geological Society of America Annual Meeting, Oct. 29, 2007, Denver, CO.

Motivation of Synthesis, With an Example on Groundwater Quality Sustainability, American Geophysical Union Fall Meeting, Dec. 12, 2007, San Francisco, CA.

The Graduate Group Model: An Innovative Structure That Is Still Maturing, Annual Meeting, ASCE Environmental and Water Resources Institute, May 15, 2008, Honolulu, Hawaii.

Climate Change, Water Resources and Deep CO<sub>2</sub> Sequestration, UCD Summer Science Institute, July 9, 2008, Davis, CA.

Groundwater Contaminant Transport and the Utility of a Subsurface Observatory, Consortium of Universities for the Advancement of Hydrologic Sciences (CUAHSI) Biennial Colloquium, July 14, 2008, Boulder, CO.

Climate Change, Water Resources and Deep CO<sub>2</sub> Sequestration, Future Farmers of America, August 26, 2008, Davis, CA.

**Keynote:** New Paradigms for Groundwater and Surface Water Interaction, HydroPredict 2008, International Interdisciplinary Conference on Predictions for Hydrology, Ecology, and Water Resources Management: Using Data and Models to Benefit Society, Prague, Czech Republick, Sept. 16, 2008.

Climate Change and Subsurface Storage, Conference Honoring Shlomo Neuman's 70<sup>th</sup> Birthday, Oct. 25, 2008, Tucson, AZ.

Subsurface Storage and Recovery: Perspective on Climate Change and Sustainability of Groundwater Quantity and Quality in CA, Joint Monthly Meeting of American Water Resources Association and California Groundwater Resources Association, Nov. 8, 2008, Sacramento, CA.

Subsurface Storage and Recovery: Perspective on Climate Change and Sustainability of Groundwater Quantity and Quality in CA, University of Concepcion, Chile, March 18, 2009.

Subsurface Storage and Recovery: Perspective on Climate Change and Sustainability of Groundwater Quantity and Quality in CA, Special webinar presentation to CA state agency Geosymposium, April 28, 2009.

Subsurface Storage and Recovery: Perspective on Climate Change and Sustainability of Groundwater Quantity and Quality in CA, Bay Area Chapter of California Groundwater Resources Association, May. 20, 2009, Berkeley, CA.

**Keynote:** The Disconnect Between Monitoring and Modeling, NOVCARE Conference, Leipzig, Germany, May 13, 2009.

## PUBLICATIONS

### Articles and Books

Fleckenstein, J.H. and G.E. Fogg. 2008. Efficient upscaling of hydraulic conductivity in heterogeneous alluvial aquifers, *Hydrogeology Journal*, DOI 10.1007/s10040-008-0312-3.

Niswonger, R.G. and G.E. Fogg. 2008. Influence of perched groundwater on baseflow, *Water Resources Research*, 44, W03405, doi:10.1029/2007WR006160.

Niswonger, R.G., D.E. Prudic, G.E. Fogg, D.A. Stonestrom and E.M. Buckland. 2008. Method for estimating spatially variable seepage loss and hydraulic conductivity in intermittent and ephemeral streams, *Water Resources Research*, 44, 10.1029/2007WR006626.

Rains, M.C. G.E. Fogg, T. Harter, R.A. Dahlgren, and R.J. Williamson. 2008. Geological control of physical and chemical hydrology in vernal pool wetlands, Central Valley, California, *Wetlands Journal*, 28(2), p. 347-362

LaBolle, E.M., Leaist, D.G., J. Gravner, G.E. Fogg and J.W. Eweis. 2008. Diffusive fractionation of isotopes in groundwater, *Water Resources Research*, 44, W07405, doi:10.1029/2006WR005264.

Eweiss, J.B., E.M. LaBolle, D. Benson and G.E. Fogg. 2007. The role of volatilization in changing TBA and MTBE concentrations at MTBE contaminated sites, *Environmental Science and Technology*, 41(19): 6822-6827.

DePaulo, D.J., F.M. Orr Jr., S.M. Benson, M. Celia, A. Felmy, K.L. Nagy, G.E. Fogg, R. Snieder, J. Davis, K. Pruess, J. Friedmann, M. Peters, N. B. Woodward, P. Dobson, K. Talamini, M. Saarni. 2007. Basic research needs for geosciences: Facilitating 21<sup>st</sup> century energy systems. Office of Basic Energy Sci., U.S. Dept. of Energy, 287 p.

Lee, S.-Y., S.F. Carle and G.E. Fogg. 2007. Geologic heterogeneity and a comparison of two geostatistical models: sequential Gaussian and transition probability-based geostatistical simulation, *Advances in Water Resources*, 30, doi:10.1016/j.advwatres.2007.03.005.

LaBolle, E.M., G.E. Fogg and E.B. Eweis. 2006. Diffusive fractionation of <sup>3</sup>H and <sup>3</sup>He in groundwater and its impact on groundwater age estimates, *Water Resources Research*, 42, W07202, doi:10.1029/2005WR004756.

Fogg, G.E. and E.M. LaBolle. 2006. Motivation of synthesis, with an example on groundwater quality sustainability, *Water Resources Research* (special forum on synthesis in the hydrologic sciences), 42, W03S05, doi:10.1029/2005WR004372.

Fleckenstein, J.H., R.G. Niswonger and G.E. Fogg. 2006. River-aquifer interactions, geologic heterogeneity, and low-flow management, *Ground Water*, doi: 10.1111/j.1745-6584.2006.00190.x, 1-16.

Rains, M.C., G.E. Fogg, T. Harter, R.A. Dahlgren, and R.J. Williamson, 2006, The role of perched aquifers in hydrological connectivity and biogeochemical processes in vernal pool landscapes, Central Valley, California. *Hydrological Processes*, 20(5), p. 1157-1175.

Knudby, C., J. Carrera, J.D. Bumgardner and G.E. Fogg, 2006, An empirical method for the evaluation of effective conductivity of binary media. *Advances in Water Resources*, 29, p. 590-604.

Purkey, D.R., W.W. Wallender, N. Islam, G.E. Fogg and B. Sivakumar. 2006. Describing near surface, transient flow processes in unconfined aquifers below irrigated lands: A deforming finite element model for heterogeneous aquifers. *Journal of Hydrology*, 330, 10.1016/j.jhydrol.2006.04.006.

Purkey, D.R., W.W. Wallender, G.E. Fogg and B. Sivakumar, 2004, Describing near surface, transient flow processes in unconfined aquifers below irrigated lands: Model application in the western San Joaquin Valley. *Journal of Irrigation and Drainage Engineering*, Nov./Dec. p. 451-459.

Weissmann, G.S., Y. Zhang, Fogg, G.E., and Mount, J.F., 2004, Influence of incised valley fill deposits on hydrogeology of a glacially-influenced, stream-dominated alluvial fan. SEPM special publication on Hydrogeophysics and Hydrostratigraphy (edited by John Bridge and David Hyndman), p. 15-28.

Fleckenstein, J.H., Anderson, M, Fogg, G.E. and Mount, J., 2004, Managing Surface water-groundwater to restore fall flows in the Cosumnes River. *Journal of Water Resources Management and Planning*, July/August, 301-310 (received Best Paper Award).

Hopmans, J.W. and G.E. Fogg. 2003. Soil water flow under saturated conditions. *Encyclopedia of Water Science*, Marcel Dekker, New York, 871-874.

Zhang, Y. and G.E. Fogg. 2003. Simulation of multi-scale heterogeneity of porous media and parameter sensitivity analysis. *Science in China (Series E)*, 46(5), 459-474.

Lu, S., F.J. Molz, G.E. Fogg and J.W. Castle. 2002. Combining stochastic facies and fractal models for representing natural heterogeneity. *Hydrogeology Journal*, 10: 475-482.

Knudby, C., J. Carrera, and G.E. Fogg. 2002. An empirical method for the evaluation of the equivalent conductivity of low-permeable matrices with high-permeable inclusions. IAHR/IAHS International Groundwater Symposium, March 25-28, Berkeley, CA.

Weissmann, G.S., Z. Yong, .E.M. LaBolle, and G.E. Fogg. 2002. Dispersion of groundwater age in an alluvial aquifer system, *Water Resources Research*. 38(10):16.1-16.8.

Weissmann, G.S., J.F. Mount, and G.E. Fogg. 2001. Glacially-driven cycles in accumulation and sequence stratigraphy of a stream-dominated alluvial fan, San Joaquin Valley, California. *Journal of Sedimentary Research Section A-Sedimentary Petrology & Processes*. 72(2): 241-250.

Fleckenstein, J., E. Suzuki and G. Fogg. 2001. Options for conjunctive water management to restore fall flows in the Cosumnes River basin, California. In: M.A. Mariño and S.P. Simonovic (ed.), *Integrated Water Resources Management*, IAHS Publication no. 272, IAHS Press, Wallingford, U.K., (ISBN 1-901502-76-7).

LaBolle, E.M. and G.E. Fogg. 2001. A Review of California's Integrated Groundwater and Surface-Water Model (IGSM). In: Seo, H.S., E. Poeter, C. Zheng, and O. Poeter (ed.), *MODFLOW 2001 and Other Modeling Odysseys – Conference Proceedings*, September 11-14, 2001, 349-355.

LaBolle, E.M. and G.E. Fogg, 2001, Role of molecular diffusion in contaminant migration and recovery in an alluvial aquifer system, *Transport in Porous Media* (Special Issue on Modeling Dispersion), 42, p. 155-179.

Fogg, G. E., S.F. Carle and C. Green, 2000, A connected-network paradigm for the alluvial aquifer system, in press, in Zhang, D. and Winter, C.L. (eds.), Theory, Modeling and Field Investigation in Hydrogeology: a Special Volume in Honor of Shlomo P. Neuman's 60<sup>th</sup> Birthday, Geological Society of America Special Paper 348, p. 25-42.

LaBolle, .E.M., J. Quastel, G.E. Fogg and J. Gravner. 2000. Diffusion processes in composite porous media and their numerical integration by random walks: Generalized stochastic differential equations with discontinuous coefficients. Water Resources Research 36(3): 651-662.

LaBolle, E.M. and G.E. Fogg. 2000. Reply to comment on "Diffusion theory for transport in porous media: Transition probability densities of diffusion processes corresponding to advection-dispersion equations" by Philippe Ackerer. Water Resources Research 36(): .

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Ruud, N.C., T. Harter, G. Weissmann, G.E. Fogg, 2000, Conditional geostatistical modeling of an alluvial aquifer system characterized by poor-quality shallow groundwater, in Proceedings of the ModelCARE '99 conference; Calibration and reliability in groundwater modelling; coping with uncertainty Stauffer, F. et al. (ed.), IAHS-AISH Publication, Vol. 265, p. 360-368.

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Weissmann, G.S. and G.E. Fogg, 1999, Multi-scale alluvial fan heterogeneity modeled with transition probability geostatistics in a sequence stratigraphic framework, J. of Hydrology 226, p. 48-65.

Weissmann, G.S., S.F. Carle and G.E. Fogg, 1999, Three-dimensional hydrofacies modeling based on soil surveys and transition probability geostatistics. Water Resources Research. 35(6). 1761-1770.

Fogg, G. E., E.M. LaBolle, and G.S. Weissmann. 1999, Groundwater vulnerability assessment: Hydrogeologic perspective and example from Salinas Valley, California. In Corwin, D.L., K. Loague and T.R. Ellsworth, Application of GIS, Remote Sensing, Geostatistical and Solute Transport Modeling. AGU Geophysical Monograph.

Fogg, G.E., D.E. Rolston, D.L. Decker, D.T. Louie, and M.E. Grismer, 1998, Spatial variation in nitrogen isotope values beneath nitrate contamination sources. *Ground Water*, 6(3), p. 418-426.

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Hoyle, B.L., K.M. Scow, G.E. Fogg and J.L. Darby, 1995, Effect of Carbon:nitrogen ratio on kinetics of phenol biodegradation by *Acinetobacter johnsonii* in saturated sand. *Biodegradation*. v. 6. p. 283-293.

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Loaiciga, H. A., Randall J. Charbeneau, Lorne G. Everett, Graham E. Fogg, Benjamin F. Hobbs, Shahrok Rouhani. 1992. Review of groundwater quality monitoring network design. *Journal of Hydraulic Engineering*, Vol. 118 No. 1, p. 11-37.

Fogg, G. E., F. J. Lucia, and R. K. Senger, 1991, Stochastic simulation of interwell-scale heterogeneity for improved prediction of sweep efficiency in a carbonate reservoir, in Lake, L. W., H. B. Carroll, Jr., and T. C. Wesson, Reservoir Characterization II, Academic Press, Inc., San Diego, pages 355-381.

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